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The Atomic Industry and Human Ecology

IV—RELAX

RELAX. DISASTER IS NOT INEVITABLE. Change is inevitable and the individual man or nation lasts longer and produces more good if not strained beyond the elastic limit. And it is common knowledge, if not universal practice to demonstrate it, that the relaxed body accepts greater strain without permanent damage than does the tense.

It has been stated here previously (September, 1949) that the dominant measure is not the hazard but the estimate of the hazard. Certainly the number of atomic bombs stocked by Russia was not increased by President Truman's announcement of an atomic explosion within the U.S.S.R. Certainly Russia's immediate ability to "deliver the bomb" was unchanged by the announcement, as was that of the United States also unchanged.

What has been changed is the tenor of public discussion about atomic energy in all its aspects. Lippman suggests now that the only agreement now possible is one essentially like the poison-gas treaty. Albert Friendly concludes that, in the present situation, no one will work on the use of atomic power for civilian purposes. Admirals of the United States Navy have been heard publicly in the beliefs they have held privately for many months, especially regarding the proper tools with which the bomb should be delivered.

So many different predictions have been made, so many different conclusions stated, that we may with great confidence anticipate a number of rights as well as wrongs. Even Einstein is reported to have concluded that World War IV would be fought with rocks, while refusing to predict the major weapon of World War III. If the peaks of disagreement and controversy among nations, now usually designated as War I, II, III, *etc.*, remain as peaks rather than smooth off into a plateau, then one of the sequence will surely be fought with an aerosol (radioactive or otherwise). The "victor" must obtain something beyond rubble for his larder or eventually go bankrupt.

True civilization may arrive if and when our wars are fought with words and ideas. Controversy and dispute among men will always be with us; these things appear to be fundamental and acceptable if governed by a majority that is capable of learning from the minority.

Sumner T. Pike, Commissioner, U. S. AEC, has recently stated the *immediate problem.* "We have immediately before us the unsolved question of whether, when and how we are to get a workable, enforceable international agreement for the control of the dangerous phases of atomic energy. . . . We

have continually with us the question of the extent of dissemination of scientific and other information to the public at home which, of course, in effect means to the world. . . . There is still time to fill gaps in our knowledge, to correct misunderstandings, to allay senseless panic, and to arouse from apathy. . . . This (burden) seems to fall not so much on our traditional educational systems but on those media of communication which reach the adult citizen; on scientists, writers, clergymen, commentators, editorialists, and perhaps most of all on those untitled citizens who are looked to in their communities as leaders of thought and opinion."

With these remarks of Commissioner Pike, NUCLEONICS is in complete agreement. It is our business to disseminate scientific and technical information in the field of atomic energy. NUCLEONICS readers are leaders of thought and opinion in their communities, both social and industrial. They have a duty to share the burden.

Chairman David E. Lilienthal, U. S. AEC, has been as forthright in his remarks made on receiving the 1949 Freedom Award. The United States "should stop this senseless business of choking (itself) by some of the extremes of secrecy to which we have been driven, extremes of secrecy that impede our own technical progress and our own defense."

Lilienthal continues to say that this country should intensify its efforts to develop the peaceful applications of atomic energy. NUCLEONICS is in complete agreement with these statements; and we flatly dispute the assertion made by some (see Friendly above) that no one will work on the use of atomic power for civilian purposes. It is quite clear to us that Russia and other nations will develop atomic power for civilian purposes irrespective of the developments in the United States.

Russia and other nations need power in more or less isolated areas where coal is not available except at great cost for transportation. The United States may debate whether its immediate need for such power is great; various persons will suggest *quite accurately* that the water problem and the clean-air problem are perhaps more vital and urgent in the United States. The fact is, and has been clear to the American Public, that strength in modern peace and war lies not alone in a stockpile of last year's tools and/or weapons, but in scientific and engineering personnel, thoroughly trained in the fundamentals of their subjects and welded into working teams through industry, business, and government alike.

The late Richard C. Tolman, to whom the world owes much for his scientific and organizational contributions, speaking at Brown University in June, 1947, said, "Keeping certain facts secret, for example in the field of atomic energy, can be temporarily important and serve a useful purpose, but can make no fundamental contribution to the prevention of evil." This temporary period must now be recognized as finished. The time has come to discuss atomic bombs as openly as we now discuss air power, naval vessels, and every other weapon of war—and to discuss atomic energy for peaceful applications with all possible vigor and forthrightness.

Note: The preceding editorials in this series devoted to the subject of "The Atomic Industry and Human Ecology" were entitled: I—Waste Disposal (August, 1949); II—On Risk (September, 1949); III—On Getting the Word (October, 1949).

Some Design Problems of Nuclear Power Plants

By WARD F. DAVIDSON
Consolidated Edison Company of New York, Inc.

IT IS SOMETIMES ASSERTED that the problems raised by the possibility of large-scale power production in nuclear power plants are entirely unique and without parallel in history. Such assertions will hardly stand the test of examination; in fact it appears that there have been other problems which may be taken as helpful analogies. One of these is the development of aircraft, and it will be of aid in the present discussion to explore this further.

The problems of flight by heavier-than-air craft had been in men's minds for many years before that time and numberless efforts had been made to solve them. The first definite proof that they were solvable came in 1903 when the Wright Brothers made their famous flights at Kitty Hawk. Once this fundamental or basic step had been taken, the scientists might have said that they were ready to retire from the scene and leave the "practical development" to the "engineers and technicians;" all that was needed, they might have added, was for the waiting public to specify the size, speed and range of the aircraft it wanted, and delivery would be forthcoming.

Of course we know that things did not work out that way, and yet that is about how many people would now deal with nuclear power. They would say: "The scientists have demonstrated that energy can be released on a large scale and under control by processes of nuclear fission; it is now the responsibility of the engineers and technicians to build large nuclear power plants and operate them to supply power at negligible cost!"

If one will read of the work of Orville and Wilbur Wright he cannot avoid the conclusion that they were not in any way underestimating the difficulties that lay ahead before large high-speed long-range aircraft could be a reality. Not that they did not appreciate the importance of what they had already accomplished, nor that they did not look to the future and dream dreams, but that they were "practical men" as well as scientists and engineers.

The basic scientific step had been taken and the validity of the underlying assumptions proved by experiment, yet this was only the starting point for a rapidly expanding scientific effort which, even today, has a long list of problems.

Although the basic engineering step had been taken, further progress in aircraft design was limited until progress had been made in other fields. For one thing, lower weight motors were needed, but before these could become realities metallurgists would have to provide alloy steels that could withstand higher temperatures, and petroleum chemists would have to provide new fuels of higher anti-knock characteristics and improved lubricants. And each of these would be of little value so long as it was only a laboratory product, so entire new industries or branches of industries became necessary. Lighter structural materials for the aircraft itself and methods for fabricating them were also necessary, and thus through a list unnecessary to labor.

Another essential was that there be ways and means for paying for these developments, for they would cost uncounted millions of dollars. As we

look back now over the years since 1903, we can see that only a part of the cost has been paid directly by aviation, even though we may take full account of enormous expenditures by government in furtherance of aircraft development, particularly for military purposes. *A very large part of the cost has been borne by other industries and other fields.* The automotive industry has made major contributions because of its concern with lighter and more efficient internal combustion engines and the fuel to run them. It has contributed, too, through the development of improved structural materials and methods for fabricating them. Even the householder has shared through his support of the aluminum industry. These are only a few examples.

This is important for our immediate discussion because it will become apparent that before large nuclear power plants can be constructed on a scale that can have a significant influence on the general pattern of our social and economic life, a number of costly collateral developments must be completed. Unless part of the cost of these can be allocated to other fields, the burden may be so large as to make it inadvisable or impossible to proceed with the development of nuclear power.

What are the design problems? In outline, the general problem is very simple. The apparent and obvious solution merely involves taking the heat released by controlled chain-reaction nuclear fission in a reactor and using it in a suitable heat engine for conversion into mechanical energy which may be used to drive an electric generator. We know that each of the steps involved in the over-all transformation of energy can be made, and the problem is how they can be fitted together to make a practical power plant. A study of that will be the immediate goal.

Basic Scientific Facts

To begin with, it is important to have in mind a few basic facts:

1. Fission of certain atomic nuclei releases large amounts of energy—roughly 4×10^{10} Btu per lb.

2. The fission reaction may be made a self-sustaining or chain reaction if certain very exacting conditions are satisfied.

3. The rate of reaction may be varied from the explosive range to any desired low rate.

4. Nearly all the energy appears as heat with only a small part as energy associated with more or less delayed radioactivity.

5. Fission is accompanied by intense radiation of lethal character.

6. U^{235}, that is, the isotope of uranium with an atomic weight of 235, is the only

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WARD F. DAVIDSON, Research Engineer, Consolidated Edison Company of New York, Inc., has an intimate knowledge of New York City below ground, on the surface, and in the atmosphere above, which comes only from long association with the many problems that must be solved by the engineers of power production and distribution to industry. From 1941 to 1946 he was "on loan" to NDRC as consultant and as Deputy Executive Officer. His various assignments included several with the Manhattan District, and he gave a paper, "Nuclear Energy for Power Production," at the Hague, 1947. He is Vice Chairman, Nucleonics Committee, AIEE and a Member, Committee on Nuclear Studies, IRE. He is active also in ASME and AMS. One of his active interests is in the study of atmospheric pollution, a field in which he has published and in which he has invented instruments. He knows and understands the problems of subsurface water as few engineers do. NUCLEONICS is proud to present this engineering analysis of nuclear power plants at this critical time in economic and social history.

—The Editor

fissionable isotope that occurs in nature; U^{233} and Pu^{239} are fissionable isotopes that may be made in reactors under proper conditions, by transmutation from thorium or from natural U^{238}.

These facts require some elaboration.

The importance of the first fact, namely the very large potential energy per unit of weight, comes chiefly from the almost weightless character of the "fuel." As compared with coal the weight ratio is one to three million, and this ignores the weight of air necessary to burn the coal. It is this characteristic, more than any other, that has caught the public fancy and it is this which gives to nuclear "fuels" much of their possible value. A "weightless" fuel, *in the absence of tariff barriers and*

TEN YEARS AGO—1939

The total energy reserves available in the U-atoms is 3×10^{16} m-kg or 7×10^{15} kwh. We can compare this figure with the total energy generated by the Reichselektrowerke, about 7×10^{5} kwh in 1929. *About four tons* of uranium should suffice to maintain the same energy output for 10^{5} hours, or about eleven years. To maintain a constant output, one would have only to cleanse the system from time to time *(for instance, once every day)* from the products of fission.

—*"Can Nuclear Energy be Utilized for Practical Purposes?" by S. Flügge*
Naturwissenschaften **27,** 402 (1939)

Assuming that U-235 would be the active ingredient if a chain reaction should be possible, we find that uranium has no very great economic advantage over coal. . . . the day of free atomic power is probably not yet in sight.

—*"Uranium and Atomic Power" by R. B. Roberts and J. B. H. Kuper,*
Journal of Applied Physics **10,** No. 9 (1939)

TODAY—1949

The gasses discharged from the stack of a coal-fired boiler present difficulties, but these difficulties are simple in comparison with the problem of the radioactive gasses emitted from a nuclear reactor. . . . If the cost of generating power were dependent only on the price of fuel, here would be a power Utopia, indeed. But the reprocessing of the fuel will involve elaborate and therefore relatively expensive chemical operations. And so, the capital costs of the chemical plant, the nuclear reactor, and the steam and electrical portions of the complete atomic power plant are bound to be the dominant portions of the cost of electrical energy at the point of generation.

—*"Prospects in Industrial Application of Atomic Energy" by Philip Sporn,*
President, American Gas and Electric Service Corporation,
at the Annual Convention of the National Coal Association
New York, October 7, 1949

The Russians are most certainly interested in bomb development and production. They are probably engaged in an all-out effort to make better bombs and more bombs, realizing that it is the possession of a large stock pile of bombs that determines their military effectiveness. But it seems most unlikely to me that this is the only aim of their atomic energy program. I surmise that they recognize in atomic energy a new field, the successful development of which on a broad front, would allow them to achieve in one jump a position of more equal industrial development with the rest of the world. . . . New developments in atomic energy can be expected to contribute both to peacetime and wartime strength without possibility of sharp distinction between them.

—*"Our Progress in Atomic Energy" by Robert F. Bacher,*
California Institute of Technology,
at Town Hall, Los Angeles, October 3, 1949

artificial price control, would have the same cost at any place in the world. Aircraft and surface vessels might be made almost free from the high price they must now pay for carrying their fuel with them.

The possibility of establishing a self-sustaining fission reaction is of vital importance. There are other nuclear reactions which release large amounts of energy, but they are not self-sustaining and none of them can now be made to operate except on a basis that requires far more energy to initiate the reaction than is released. (It is too much like heating a boiler with matches.) The very exacting conditions that must be satisfied pose the most difficult problems to be solved.

For power generation the explosive reaction is of little interest beyond having the assurance that the danger of accidental explosion can be made acceptably small. This is relatively easy to do. Instead of reaction times measured in microseconds and temperatures measured in millions of degrees, the power plant designer is interested in nearly steady-state conditions and temperatures no higher than that of ordinary chemical combustion.

Since most of the energy appears as heat the temperature will depend on the balance between the rate of reaction and the rate at which heat is removed. Somewhat in contrast with chemical combustion, the rate of reaction is essentially independent of the temperature within the range of engineering interest. However, there is a delayed "after effect" caused by the energy release by radioactivity which may be important in some operating and control problems, for there is no way in which this energy release can be regulated.

The radiation of alpha and beta particles, neutrons and gamma rays that accompanies fission creates a serious hazard, for in any power reactor the intensities are such as to be almost instantly lethal to any living thing. Elaborate and usually very massive shields are necessary to provide protection for personnel and to limit the zones that may become radioactive, for many materials on exposure to neutron radiation, in particular, become radioactive, thus creating the need for further shielding or protective measures.

The U^{235} isotope occurs in nature as only 0.7% of natural uranium. Under suitable conditions U^{235} may be used to transmute an approximately equal amount of the predominant U^{238} into neptunium which changes, by radioactive processes, into another fissionable isotope, plutonium-239. If the Pu^{239} is next used to initiate the transformation of more U^{238}, a "breeder" reaction has been established. In theory, it should be possible to repeat this breeder cycle indefinitely until all of the U^{238} has been converted into fissionable isotopes. A somewhat analogous transformation of thorium-232 into fissionable U^{233} may also be initiated by U^{235}. The hope of large-scale power production rests heavily on the possibility of practically utilizing U^{238} and Th^{232} in this manner.

Basic Engineering Problems

The preceding facts have been demonstrated in various reactors and processes used for the manufacture of bomb materials, notably at the Hanford Engineering Works. For various reasons, however, the temperatures in these reactors are comparatively low and the heat could be used in a heat engine only with very low efficiency.

Carnot's principle teaches that there is an upper limit to the efficiency that may be attained by any simple heat engine and that this may be expressed as

$$\eta = 1 - (T_2/T_1)$$

where T_1 and T_2 are the initial (or higher) and final (or lower) temperatures of the working fluid. In any

practical heat engine the actual efficiency will be lower than this ideal because of stray heat and mechanical losses. Under favorable conditions, with cooling water for condensers available, T_2 may be in the order of 535° R (75° F); with air cooling, as with some gas turbines, it may be near 750° R (290° F). The urge to go to higher initial temperatures is therefore evident, and considering only the heat engine, T_1 probably would approach 1,000° F for steam turbines, and 1,300° F or, possibly, 1,500° F for gas turbines.

It becomes clear, then, that for practical power generation the heat must be available at a turbine or heat engine at a temperature of at least 300° F. Several preliminary studies that have been made have been based on values in the range 750° F to 1,200° F.

We return now to consideration of the nuclear reactor itself. The first energy transformation will take place when U^{235}, or one of the other fissionable materials, undergoes fission releasing energy as heat. The fission of an atom is initiated by the entry of a neutron—an uncharged particle—into the nucleus of the atom, thus creating an unstable condition which results in "fission" into two parts and the ejection of from one to three neutrons. Since the total mass after fission is less than the original mass, the difference or loss of mass appears as energy. This follows from the Einstein $E = mc^2$ principle. The fact that some neutrons are ejected is of vital importance, for this provides the means for initiating additional fission and establishing a chain reaction. However a steady-state chain reaction requires that for every fission there shall be, on the average, one neutron which initiates a new fission. If the number is less than one, the reaction will die out; if it is greater, it will increase. The rate of change depends on the amount by which the average differs from unity, and in any power reactor this will be an exceedingly small fraction. It has been noted before that the rate of reaction is not influenced by ordinary engineering ranges of temperature, except as it influences the density of the materials in the reactor, and in this it does not have the self-regulating characteristic of many flames. At the same time the number of neutrons ejected was, on the average, less than three, so the supply is by no means unlimited. Also the supply of neutrons may be used up in many ways besides causing fission. Neutrons may escape from the reactor; they may enter non-fissionable nuclei; and they may even enter fissionable nuclei in such a way that fission does not follow.

The imperative need to keep neutron losses within tolerable limits leads to some of the most drastic limitations on freedom of design. While the designer of fuel-burning steam generators may concentrate all capacity in one large unit or divide it between several smaller units, and while he may use a tall, narrow shape or a more nearly cubical one or some other shape, the designer of a nuclear power reactor has relatively little choice.

The need to limit neutron loss by escape from the reactor leads to a spherical or cubic shape and one large unit rather than several small ones, for only in these ways can an acceptable surface-to-volume ratio be obtained. Some help can be had through the use of neutron reflectors, but at best this is not great.

Mere increase in volume is ineffective, however, if it is accomplished by the addition of inactive materials and a corresponding reduction in the average "density" of the active charge. All materials absorb neutrons to a greater or less extent, and the designer must exercise great care in the selection of materials to avoid those showing high specific absorption (large neutron-capture cross section) and then to keep the

quantity to the lowest amount consistent with the satisfactory performance of the intended function.

These requirements have been stated in such general terms that their significance may not become fully apparent until a number of somewhat more specific requirements have been examined.

The control system must be sensitive with rapid response and yet be designed to give stability and to insure an unusually high degree of reliability. To meet these requirements several new sensing elements or primary detectors must be devised. These in themselves present challenges to the instrument designer, the more so because in some cases the very activities that it is essential to measure are destructive of the instruments. Additionally, the control system itself presents several conflicting requirements arising from such factors as the wide range of time-constants and response characteristics. In fact, in a recent paper Cochran and Hansen* express the opinion that finding a good solution to these problems may be crucial to the future of nuclear power.

The materials used in the active zone of the reactor must not undergo significant changes in their essential characteristics as a result of exposure to radiation. While mechanical engineers have become accustomed to such concepts as fatigue and creep of metals and recognize the problems of corrosion, the introduction of nuclear reactors brings in a whole group of new problems. Under the intense bombardment by all types of nuclear particles that occur in the active zone of a reactor, no engineering materials are entirely immune to change, and the first task is to insure that the changes which do take place are within tolerable limits.

The engineer venturing into this field faces two handicaps. One is the almost complete lack, until very recently, of laboratory facilities for subjecting test specimens to radiation of sufficiently high intensity to permit reducing exposure times to convenient values. The other is the great complexity that results from the fact that many engineering materials are alloys whose desirable characteristics are usually much more sensitive to changes in some of the minor alloying elements than to

THE POWER RATINGS of most of the operating piles are measured in kilowatts or even megawatts. For example, the air-cooled graphite-uranium piles at Oak Ridge and Harwell, England (BEPO, the larger of the two reactors there), have been reported to operate at greater than 2000 kw and about 6000 kw, respectively. The power rating of the water-cooled Hanford piles (whose chief function is plutonium production) has not been released but is certainly much larger than that of the Oak Ridge pile. The first nuclear reactor built in 1942 in Chicago and later reconstructed at the Argonne Laboratory near Chicago consists of uranium and uranium oxide lumps in a graphite lattice with no cooling provisions and has been reported to operate at a few kilowatts. The water-cooled Los Alamos water boiler has operated at about 10 kw. The heavy-water reactor at Argonne (in which the heavy water also serves as a coolant) has been reported to run at a power level of 300 kw.

—Gerhart Friedlander and Joseph W. Kennedy, "Introduction to Radiochemistry" (John Wiley & Sons, Inc., New York, 1949)

* D. Cochran, C. A. Hansen, "Instrumentation for a Nuclear Reactor," NUCLEONICS 5, No. 1, 4 (1949).

changes in the basic element. For instance, high-strength aluminum alloys show ultimate tensile strengths more than twice that of commercially pure aluminum, and yet the total of all the alloying elements may not exceed six or seven percent. In such cases the factor of greatest importance may be, for example, the effects of radiation on the silicon content and disposition rather than the effects of radiation on the aluminum constituent. Every engineering material, and each variant of that material, presents its own list of problems, so the total task is one of impressive complexity.

Equally important with the requirement that structural materials be stable under radiation is the requirement that the specific absorption for neutrons be low. The need for limiting the loss of neutrons so that there will be enough to sustain the chain reaction has been pointed out. Unfortunately most common engineering materials must be given a poor rating on this basis and there is need for extensive investigations to find acceptable materials.

Another requirement is that there be some heat transfer medium that can absorb the heat released in the reactor and make it available for use in the turbine or other heat engine. Two considerations, in particular, make this difficult to satisfy. For one, the needs for stability under radiation, low specific neutron absorption, and high specific heat are conflicting in some degree and not satisfied by any common fluid. For the other, the materials that most nearly meet the first requirement cannot be used directly in a heat engine. A two-stage system seems to be almost a necessity. Other requirements such as thermal stability and noncorrosiveness only add further complications.

Adequate shielding for the protection of personnel and equipment against radiation is another essential. Basically it can be satisfied by providing a shield of sufficient mass to absorb the radiations or attenuate them to acceptable levels. Practically, it involves a number of difficult problems as how to provide the necessary access doors and openings and how to avoid structural change and deterioration. It may also require the use of two heat-transfer media with a heat-exchanger that is also a radiation shield between them.

First Outline of a Power Plant

With this background, some attention now may be directed toward exploring possible nuclear power plant arrangements.

Of the various heat transfer media only two seem to offer enough possibility of meeting the requirements set by the reactor and also to be usable directly in a heat engine. These are air and helium. Both could be used in a gas turbine, but it is noted immediately that air would be radioactive and both might become contaminated with radioactive fission products. Consequently it seems almost mandatory to provide for a heat exchanger which will serve to isolate the radioactivity and keep it from the turbine.

Once this has been done, all the media come on a common basis as requiring a heat exchanger or steam generator, inasmuch as neither water, molten cadmium, or molten lead can be used directly in a heat engine.

For the heat engine, a steam cycle offers by far the greatest degree of flexibility, as the upper temperature may be almost anywhere in the range 250° F to 1,050° F with the possibility of even higher values. The range of upper or initial temperatures for gas turbines is much more limited—say, from 950° F to 1,500° F on the basis of present-day engineering developments. As explained before, the higher temperatures offer attraction in the way of improved conversion efficiency, but they also lead to serious difficulties in the

design of the reactor. It probably will be many years before these can be overcome sufficiently to permit utilizing the full possibilities of the gas turbine.

The power-plant design thus has been narrowed down to a reactor transferring heat to a fluid heat-transfer medium which is circulated through a steam generator to produce steam which is utilized in a turbine driving an electric generator. The turbine and generator can be quite conventional and will require no further attention. The steam generator presents some new, but not unique, design problems. In particular, more than usual care will be needed to develop a construction which will be free from leakage to insure against contamination of the steam by radioactive materials. It will be necessary, too, to devise means for cleaning the water tubes from time to time without dangerously exposing the personnel to radiation, for it must be assumed that water contamination cannot be avoided entirely.

The most difficult, and very new, problems come in the reactor design for, as will be seen, the other power plant components do not depart so far from practice in fuel-burning stations. Whether a "homogeneous" or a "heterogeneous" reactor is chosen is not a matter of great importance for the present discussion, for the specific problems will differ in detail rather than kind. Since the "heterogeneous" reactor of the general type used at the Hanford Engineer Works has been referred to in other discussions of nuclear power plants, it will be used as the starting point for the next stage of this analysis.

In this pile the uranium is encased in close fitting aluminum tubes which are then inserted into tubes of slightly larger diameter disposed in the carbon moderator. The cooling water is circulated through the annular spaces between the tubes. As these piles were designed for plutonium production and not for power generation, the temperatures in the cooling water were kept low.

At first thought it might seem an easy step merely to reduce the flow rate for the water until the temperature rose to the boiling point and steam was generated. A further flow reduction could result in delivering superheated steam. There is however an immediate and major objection. It arises from the fact that, particularly at low pressures, the density of steam is much less than that of water and consequently the neutron absorption coefficient will be correspondingly less. This, in turn, will cause disturbances in the nuclear reactions in the pile which may become so violent as to lead to instability. If the pressure were increased to the critical value (3,206 psi) this trouble would be avoided, but other troubles would be introduced through the need for far heavier tube walls to withstand the higher pressures. Additionally, the higher steam temperature (705° F) would exceed the safe working temperature of aluminum. The introduction of more structural metal into the reactor, as would be necessary at the higher pressure, would have to be examined critically, as would the total amount of heat-transfer fluid within the reactor, for both have an adverse effect on the neutron absorption and the performance of the reactor.

An evaluation of heat-transfer fluids presents a complex and difficult problem because of the many factors that are involved. Gilliland has made an interesting start.* Taking into account various physical properties such as specific heat, thermal conductivity, density, viscosity, and the power required to circulate the fluid, he has developed two sets of parameters which measure the relative merits of different

* "The Science and Engineering of Nuclear Power," chapter on *Heat Transmission* by E. R. Gilliland (Addison-Wesley Press, Cambridge, Mass., 1947).

fluids. He calculated the values of these parameters for 21 gases and vapors and for 15 liquids and molten metals. A study of these values and of data on neutron capture cross section led him to select four media for further study. They are helium, water (in spite of the objections noted above), molten cadmium and molten lead. Other investigators have added air and some eutectic mixtures involving lead.

This list includes several fluids that have been used but little as heat transfer media, and not at all at high transfer rates. Before any of these can be used, extensive studies of heat-transfer performance will be necessary, and these are engineering studies of great complexity.

Some Problems of Materials and Processes

Metals suggest themselves at once as the principal structural materials, although ceramics offer some interesting possibilities. Of the metals, low alloy steels seem to be excluded on several bases: high neutron absorption, low strength at elevated temperature, questionable stability under radiation. Aluminum, although used in the reactors at Hanford Engineer Works where the temperatures are quite low, seems clearly out of the picture for power reactors because of low strength at high temperatures. Some of the stainless irons, such as "18-8" stabilized austenitic, have possibilities although not wholly desirable on the basis of either neutron absorption or thermal conductivity. The examination might be extended through the whole list of metals commonly used in engineering and none would be found not open to major objections. This suggests examining some of the higher melting point metals that have not been used for engineering purposes—beryllium, titanium, molybdenum, tantalum, wolfram —to name a few. The metallurgy of these metals has not been developed sufficiently to permit drawing conclusions at this time. It is certain that any of them would be costly, if not prohibitively so. The physical properties of these metals when fabricated in sizes suitable for power plant use are not well known. At best, these materials should be classed as "interesting possibilities."

Ceramics have been suggested, but it would be misleading to consider them as direct substitutes for metals, because their interesting and valuable properties can be realized only when the basic machine design is developed with their use in mind. This, in turn, requires development work in new fields of engineering, detailed study and analysis.

If we can assume that a tentative solution has been found to the heat transfer problem by itself, then we may pass on to several others.

Some means must be devised to permit removing fission products and replenishing the supply of fissionable materials. As the range is not large between the lower limit of density of fissionables necessary to maintain the chain reaction and the higher limit of density when control becomes difficult, the maintenance of satisfactory operating conditions requires an approach to a continuous process. If we continue to refer to the Hanford-type piles, this means the removal of one bar or slug of uranium in its aluminum case and its replacement by another.

A continuous process of some type, rather than a batch process, would have many advantages. As the continuous fuel feeding of a stoker or a powdered coal burner has replaced intermittent hand firing, and as continuous ash or slag removal has replaced intermittent grate cleaning in coal-fired boilers, so continuous feeding and purging may be expected to be developed eventually for reactors. Just how this can be done is not now clear, for the difficulties are not alone those of a mechanism to handle

Underlying all aspects of a nuclear power plant are the extreme precautions that must be taken to assure that there be no uncontrolled escape of dangerous radioactive substances and that structural components that become radioactive either last for an acceptable plant life time or be made susceptible to replacement by remote control.

—I. Perlman, "The Outlook for the Nuclear Power Plant," presented at AIEE Pacific General Meeting, August, 1949

the radioactive materials, but even more those of designing a reactor which lends itself to such processes.

Special tools for maintenance and repair will have to be devised, for the presence of radiation, even when the reactor is not operating, will make it unsafe to use the usual tools and techniques of conventional fuel-burning power plants.

Although not truly a part of the reactor and the power plant, provision must be made for the treatment of the depleted charge removed from the reactor. The need for conserving fissionable materials, especially if nuclear power generation is to become widespread, is such as to necessitate the careful processing of the charge as removed from the reactor to separate the unspent fissionable materials from the fission products. The unspent materials would then be reprocessed and used again while the fission products would be disposed of. This will probably be chiefly as waste materials, although it seems probable that a few of the constituents will have other uses which would justify the costs of recovering them. The safe disposal of the unusable wastes presents further problems. These radioactive materials cannot be dumped in ordinary refuse piles, but must be buried on land or disposed of far at sea in such ways that they will not bring death to animal or plant life.

The need for adequate shielding has been mentioned. This is not peculiar to reactors used for power generation; experience being gained in the design of research reactors should provide most of the needed information.

The problem of controlling a reactor so as to maintain some desired level of activity has been well discussed by Friedman.* For a power unit, however, where the usual requirement is to meet some power output demand, the problem is considerably more complex. In fact, it may be so great that it will be necessary for the first installations to be operated under conditions that will permit disposing of whatever power the unit may tend to generate at the time by feeding the power into a system having other generating capacity which can absorb the variations.

There are many other details that will need attention before a practical nuclear power generating unit can be built. Few pieces of auxiliary equipment, such as fans, pumps, heat exchangers, flow meters, and many others that are common in fuel burning power plants, the heavy chemical industries and factories generally, can be used without more or less extensive modification. The change may involve the substitution of materials less susceptible to change by radiation, or major redesign so that when the inevitable repair or replacement becomes necessary the work can be done safely by the use of special tools and devices. In other cases a wholly new design may be needed.

Some General Problems

The discussion up to this point has considered the more general engineering problems for which some solution must be found before a nuclear power plant of engineering significance can be built.

* F. L. Friedman, *Electrical Engineering* **67**, 685 (1948).

If we can judge by the experience in other fields, it is not to be expected that the first units—the pilot plant units—will show great similarity with units that may be built after 10 or 15 years of experience, of trial and error.

Nonetheless the broad general outlines are, perhaps, now sufficiently clear so that it may not be venturing too far into the realms of fantasy and wishful thinking to look at some of the economic factors that are involved. In doing so it must be kept in mind that there are special applications, chiefly military and naval, when considerations of expediency in particular values outweigh economic considerations. But in general nuclear power will not become widely useful unless it can be developed to a point where it can justify itself on a cost basis.

It will help the analysis to take a conventional fuel-burning steam turbine power station as a starting point and note similarities and differences.

No important change is required in the turbine-generator unit with its condenser, nor in associated switchgear.

The steam generator will differ in several details from a fuel-burning unit. If the heat-transfer fluid is a gas, the unit may have many of the characteristics of a waste-heat boiler; if the heat-transfer fluid is a liquid or molten metal, it will take on quite new characteristics. In either case the heat transfer will be predominantly by conduction and convection and almost not at all by radiation. This will have a marked influence on the design, especially in the steam generating sections. More uniform use of the heat transfer surfaces may be expected; taken with the elimination of the furnace volume, this will result in significant reductions in size of the steam generator. Structurally it will be necessary to place particular emphasis on tightness, for any leaks might result in contamination which might be troublesome on either side. This may require the use of corrosion-resistant alloys and almost complete welding of all joints. New provisions for cleaning and purging the steam side will be necessary, as it must be possible to carry out these operations without exposing the operator to dangerous radiation. If molten metals are used for heat transfer, some means must be provided for draining the metal on shutdown of the reactor or for melting it in place when starting up.

The reactor and the fluid heat-transfer system have no close counterpart in the fuel-burning station. They are entirely new elements. In bulk they probably will be rather smaller than the furnace combustion space for an equal output: Gilliland, in one example, has assumed a reactor in the form of a cube ten feet on a side as appropriate for a 45,000-kw unit. To this must be added the heavy radiation shield of concrete or similar material and space for the fluid circulating pumps. There is no need to discuss again the general requirements for feeding fissionable materials and removing fission products. The control system quite certainly will be more complex than for the fuel-burning station.

The nuclear power station will not be required to handle large weights of fissionable materials or fission products. On the other hand the radioactivity of the fission products will require elaborate, and probably bulky, equipment to permit safe handling. Cranes may be larger than the coal hoists replaced.

The nuclear station that we have pictured will not require forced and induced draft fans, nor air preheaters, but there will be other auxiliary equipment such as circulator pumps of possibly equal bulk and about equal complexity.

If the reactor can be so designed that the fissionable materials, and the fission products, are tightly encased in metal capsules which are not opened until delivered to a chemical reprocessing

and recovery plant, then the quantity of radioactive gaseous products to be handled at the power plant will be limited and comparatively small, and simple facilities will be adequate for their safe disposal. However, this does not obviate the necessity for facing the problems of disposing of the radioactive wastes; it merely shifts them to the chemical plant.

The picture of a nuclear power plant that thus emerges shows a turbine room and electric equipment galleries quite like those in conventional coal-burning stations, although with smaller but much more complex equipment where the boiler room and coal handling equipment had been.

Economic Considerations

What will such a plant cost, and what are the prospects that it can produce energy at a cost that will be attractive economically? One would be venturesome indeed to attempt to answer that question in view of the lack of available data as to probable costs of the essential nuclear fuels. About all that can be said is that plant costs will probably be considerably higher than for fuel burning installations. The "fixed charges" will be correspondingly higher. In this respect they will be more like hydroelectric installations.

They may also be more like hydroelectric than fuel-burning stations in that the cost of "fuel" (which is often considered nil for water power) may be much less, relatively, than for coal-burning power plants.

A few further comments may be added. In making a comparison of nuclear and fuel-burning power plants, it should not be assumed that they can be placed on sites equally advantageous with relation to the load. Until years of successful operation have proved their reliability, no prudent engineer would place a large nuclear power station in the heart of a large city or in densely populated or industrially very important areas. The ever present chance, however small, that something might go wrong and release large amounts of radioactive material would dictate sites in areas where a failure would be less disastrous. Also, as mentioned before, the problem of safe disposal of radioactive products may raise problems that are solved much more easily in outlying areas. Such considerations may lead to significant increases in transmission costs, which would offset some of the possible advantages of lower generation costs.

Although it frequently has been suggested that nuclear power plants offered particularly attractive advantages in areas where adequate supplies of condenser cooling water were not available in lakes or rivers, this does not meet the test of critical analysis. As a heat engine—a steam turbine or a gas turbine—is required in either a nuclear or a fuel-burning plant, the conversion efficiency will be influenced equally by the availability or lack of cooling water. Higher final temperatures mean lower efficiency, larger turbines, larger steam generators, larger reactors, and so forth, and higher fuel consumption. All that can be said with certainty is that at some time in the future nuclear power plants may not be influenced as adversely as fuel-burning power plants by lack of natural cooling water supplies.

The last part of this discussion applies specifically to power plants for central power system supply and, quite clearly, it is not applicable to a number of other cases. For example, for ship propulsion, and especially in naval vessels, the possible savings in weight of "fuel" may offset a large increase in first cost. The hazards may be minor in comparison with others that are accepted. Each case involves distinct problems. It would be folly to argue from one to the other without checking the validity of the assumptions. —END

Place in Periodic System and Electronic Structure of the Heaviest Elements

Historical Background

Before the Discovery of the Transuranium Elements. The intensive study of the heaviest elements during the last few years has given information and data which now enable us to make some definite statements on their electronic structure and their place in the periodic table. These are to an appreciable extent two different questions because, as for example in the case of the rare-earth elements, such elements will probably find their place in the periodic system on the basis of their chemical properties as a group rather than on the basis of the individual electronic structures of their gaseous atoms. The information obtained about the recently discovered synthetic transuranium elements has been particularly useful in this connection, and it is largely on the basis of these new elements that this question is now well understood.

The heaviest natural elements, thorium, protactinium and uranium, of atomic numbers 90, 91 and 92, respectively, have been placed in corresponding positions just below the 6th period "transition" elements, hafnium, tantalum, and wolfram, in which the 5*d* electron shell is being filled. The elements, hafnium, tantalum, and wolfram are similar in their chemical properties to the corresponding transition elements in the 5th period, zirconium, niobium, and molybdenum, in which the 4*d* shell is being filled.

It has long been known that the chemical properties of thorium, protactinium and uranium resemble somewhat those of these 4*d* and 5*d* elements, and for this reason most of the textbooks and standard works on chemistry and physics in which the electronic structure is discussed have accepted the view that it is the 6*d* shell which is being filled. Thus the structure of the elements above radon (element 86) through uranium has been written to show the addition of the next two electrons in the 7*s* shell for element 87 (francium) and element 88 (radium) and addition in the 6*d* shell for the following four elements, actinium, thorium, protactinium and uranium (*1*). Many of the early papers which appeared after Bohr's classical work (*2*) on the quantized nuclear atom discuss the electronic structure of the heaviest elements. It has been recognized by many that the next hypothetical rare gas should have the atomic number 118, in a place 32 elements beyond radon ($Z = 86$), thus implying transition groups similar to those between xenon ($Z = 54$) and radon ($Z = 86$). Rydberg (*3*) as early as 1913 implied that this was to be expected. There has been general agreement that some type of transition group should begin in the neighborhood of uranium, although there has been difference of opinion as to where it begins and which electron shell is involved.

A number of the earliest publications even have suggested that this transition series involves the filling of the 5*f* shell, thus possibly giving rise to a "rare earth" group in a manner analogous to that resulting from the filling of the 4*f* shell. The filling of the 4*f* shell results in the well-known group of 14 rare-earth elements of atomic numbers 58–71 inclusive, following lanthanum. It is

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of interest here to note a few of these early and also later suggestions in order to review the general previous status of this question. Most of these early investigators were of the opinion that the filling of the 5*f* shell should begin at some point beyond uranium, that is, beyond the then known elements.

In early papers Bohr (*4*) suggested that the addition of the 5*f* electrons might begin in this region, and in a Bohr-Thomson type of periodic table he pictured the first entry at the element with atomic number 94. Goldschmidt (*5*) felt that the transuranium elements up to atomic number 96 should be homologues of the platinum group and Hahn (*6*) felt that this view was worthy of serious consideration. Suguira and Urey (*7*), using the old quantum theory, published the results of their calculations indicating that the first entry of an electron into the 5*f* shell should occur at element 95, while Wu and Goudsmit (*8*) on the basis of a more refined calculation showed that their solution of the Schrödinger equation indicated such entry at uranium or element 93. McLennan, McLay, and Smith (*9*) suggested as an alternative to the filling of the 6*d* shell the possibility that the 5*f* shell begins to be occupied in thorium. In a review article, Dushman (*10*) stated that it is doubtful that the added electrons enter the 6*d* level (thus implying an analogy with cerium, *etc.*). Swinne (*11*) pointed out that the available evidence on thorium and uranium was consistent with first entry of 5*f* electrons at protactinium or uranium but also thought that it might occur beyond uranium. M. N. Saha and N. K. Saha (*12*) suggested, as an alternate possibility to the filling of the 6*d* shell, the entry of the first 5*f* electron at thorium. Karapetoff (*13*) suggested that the element with atomic number 93 might be the first in which the 5*f* shell begins to be filled, while Grosse (*14*) suggested, as a possible alternative to filling of the 6*d* shell, the entry of the first electron in the 5*f* shell at uranium. Quill (*15*), largely for the purpose of illustration, presented periodic-table arrangements in which the first 5*f* electron appears in element 95 in one case and in element 99 in another. Perrin (*16*), Rudy (*17*), and Carranza (*18*), on general considerations, proposed as a possibility the theory that the first 5*f* electron appears in thorium and Villar (*19*) more recently suggested that some of the chemical evidence supports this viewpoint of an "actinide" transition group. Somewhat earlier, on the basis of his crystallographic work, Goldschmidt (*20*) had changed his original point of view and came to the view that the first 5*f* electron enters at protactinium, the first element beyond thorium, although he pointed out the possibility that this might occur either earlier, in thorium, or later, in uranium or in the (at the time unknown) transuranium elements. By analogy with the name "lanthanide" series which he had already proposed (*21*) for the rare-earth elements because these 14 elements following lanthanum have lanthanum as their prototype, he proposed the name "thoride" series for the 14 elements following thorium. At the same time he was the first to suggest that terms such as "actinide," "protactinide" and "uranide" might describe the group should it eventually be found to begin earlier or later than this.

It can be seen that, although many interesting and perspicacious proposals had been made up to this time, the

electronic structure and place in the periodic table of these elements could not be regarded as established.

After the Discovery of the Transuranium Elements. The recent identification of the transuranium elements [following Hahn and Strassmann's (*22*) famous discovery of the fission process] and the study of their properties, especially the chemical properties, have given us a tremendous amount of additional evidence of just the type needed to clarify this problem.

As it turns out, it is in the transuranium elements that the really definitive chemical properties, from the standpoint of placing the heaviest elements in the periodic table, first appear. The first good definite evidence that the 5*f* shell undergoes filling in this heavy region came from the tracer chemical observations of McMillan and Abelson (*23*) on element 93 (neptunium); upon their discovery of this, the first transuranium element, they were able to show definitely that it resembles uranium in its chemical properties and bears no resemblance to rhenium, the element immediately above it in the periodic table. This excellent experimental evidence was interpreted by them to indicate that this new "rare-earth" group of similar elements starts with uranium. The later calculations of Mayer (*24*) indicate that the energy and spatial extension of the 5*f* eigenfunctions drop sharply at about element 91 and therefore that the filling of the 5*f* shell might begin at protactinium or uranium. Daudel (*25*), in commenting on this paper, made some remarks concerning the special nature of the *f* electrons at the beginning of the rare-earth and the heavy transition groups. Starke (*26*) and Bedreag (*27*) also interpreted the tracer experiments with element 93 as indicating that the first 5*f* electron comes at element 93, while Strassmann and Hahn (*28*) felt on the basis of their tracer experiments with this element that it was difficult to make any deduction. As a result of their first tracer experiments with element 94 together with their consideration of the tracer investigations with element 93, Seaborg and Wahl (*29*) in 1942 made the suggestion that this transition group might begin earlier and that thorium or actinium might be the zero element in the series. On the basis of his complete crystallographic evidence, including especially observations on the transuranium elements, Zachariasen (*30*) felt with Goldschmidt that a "thoride" series is involved and the latter (*31*) felt that the chemical properties of neptunium strengthened this point of view. Smyth (*32*) in his famous report on the Manhattan Project suggests a uranide series. Some spatial classifications (*33*, *34*) of the elements have appeared recently in which the heaviest elements, starting with thorium as the homologue of cerium, are listed as the chemical homologues of the rare-earth elements, but the reason in these cases appears to be mainly connected with the symmetry of, and the ease of making, such an arrangement.

Following the first tracer work on the transuranium elements, the elements neptunium (atomic number 93) and plutonium (atomic number 94) have been extensively investigated with substantial, weighable quantities. Americium (atomic number 95) and curium (atomic number 96) also have been available for investigation, on a more limited scale, in weighable quantities. The recent extensive investigations on thorium, protactinium, and uranium also have contributed to the evidence which is now useful to interpret this question. All of this information, and especially the chemical evidence concerning americium and curium, seems to lead to a self-consistent and unambiguous picture concerning the nature of this transition series.

Actinide Concept

General. The evidence now available leads to the definite view that it is the 5*f* electron shell which is being filled in these heaviest elements. The evidence seems sufficient to go farther than this and suggest (*35*, *36*) that this rare-earth-like series begins with actinium in the same sense that the rare-earth or "lanthanide" (*21*) series begins with lanthanum. On this basis, it might be termed the "actinide" series (*35*, *36*), and the first 5*f* electron might appear, although not necessarily, in thorium. The salient point is that the characteristic oxidation state (*i.e.*, the oxidation state exhibited by the member containing seven 5*f* and presumably also by the member containing fourteen 5*f* electrons, curium and element 103) is the III state.

There is much evidence, all pointing toward this view. It will be discussed in relation to: (1) chemical properties, (2) absorption spectra in aqueous solution and crystals, (3) crystallographic structure data, (4) magnetic susceptibility data, and (5) spectroscopic data.

It should be emphasized that the discussion so far has been carried on in a somewhat oversimplified manner, because the details concerning the possible physical or chemical forms in which these elements might exist have a bearing on the electronic structures, as is the case for all the other transition groups including the rare-earth elements. Differences of this sort should be much more pronounced for the 5*f* than for the 4*f* transition group because of the lower binding and shielding of the 5*f* electrons. The effect which this should have on making the heavy "rare-earth-like" series different from the light rare-earth group has been generally not well recognized, even though Bohr (*37*) pointed out clearly more than twenty-five years ago that this looser binding would obscure the place of beginning for the heavy series. Thus the number of 5*f* electrons in the atom in the gaseous state might differ from that in the metallic state (which in turn can differ from one phase to another), and in turn neither of these structures might correspond directly to the chemical properties, in which hydration and lattice energies play an important role. It is the chemical properties (including absorption spectra, crystallographic data, *etc.*, on the compounds) which should be determinative in placing these elements in the periodic table, just as was the case for the rare-earth elements which would be placed differently considering their electronic structures alone.

Chemical Evidence. On the basis of an actinide series, the characteristic oxidation number for the series is III, and this shows up strikingly in the stabilization of the lower oxidation states with increasing atomic number. In going from uranium to plutonium, it becomes increasingly difficult to effect the oxidation from the IV to the VI state and, in fact, with americium it is impossible in aqueous solution to effect an oxidation to the VI state. Similarly, it becomes increasingly difficult to effect oxidation from the III to the IV state in going from uranium to plutonium, and with americium the evidence indicates that it probably is not possible to effect this oxidation in acid solution at all.

TABLE 1

Some Oxidation Potentials of the Actinides

(*Aqueous Solution, 1 Molar*)

Element	*Potential in Volts* III–IV	IV–VI
92-U	+0.63	−0.33
93-Np	−0.14	−0.94
94-Pu	−0.95	−1.0
95-Am	~ −2.6	

If this oxidation should be proved possible, the indications are that the potential is so great that the higher oxidation state of americium is reduced by water and cannot be maintained in aqueous solution for any great length of time. These considerations are illustrated by Table 1, in which the standard oxidation-reduction potentials, referred to the hydrogen-hydrogen ion couple as zero [see, for example, W. M. Latimer, "Oxidation Potentials" (*38*)], are listed for the III–IV and IV–VI oxidations for these elements (*39*).

Much of the work done with curium has of necessity been limited to the tracer scale; therefore it has been impossible to make corresponding quantitative deductions. This work, however, has led to the definite qualitative conclusion that it is impossible in aqueous-acid solution to oxidize curium to the VI state and that it is also impossible to oxidize it from the III to the IV state. In fact, the experiments of Thompson, Morgan, James, and Perlman (*40*), in which tracer amounts of curium and americium were subjected to strong oxidation under alkaline fusion, indicate that it is more difficult to oxidize curium from the III to an upper state than is the case for americium, and it may be impossible to effect this oxidation at all. These experiments indicate that americium may be oxidized in alkaline media, and can in this manner be separated from curium. In fact, Werner and Perlman (*41*) were able to oxidize americium (III) in 40% potassium carbonate solution by the use of the strong oxidizing agent hypochlorite to an insoluble compound, probably of americium (V), and hence to effect an efficient separation from curium (III) which apparently is not oxidized under these severe conditions. The microchemical experiments of Werner and Perlman (*42*) with macroscopic concentrations of curium also point toward the existence of curium solely in the III oxidation state in aqueous solution.

This tendency toward increasing stabilization of the lower oxidation states, especially the III state, with increasing atomic number also manifests itself notably in the stability of the solid compounds of the various oxidation states of these elements. The best illustration arises from a consideration of the solid non-oxygenated halides of these elements. The first possibility of the production of a trifluoride appears with uranium trifluoride which can be prepared only under drastic reducing conditions, and the stability and ease of reduction to the trifluoride increases in going to neptunium and then to plutonium. In the case of americium, it has not been possible to produce any higher fluoride than the trifluoride.

With respect to the other halides, it has, in fact, been impossible to prepare any plutonium or americium chloride, bromide, or iodide of oxidation state higher than III, and it has been possible

TABLE 2

Halides of Some of the Heaviest Elements

Element	*Fluorides*				*Chlorides*				*Bromides*		*Iodides*	
92-U	UF_6	UF_5	UF_4	UF_3	UCl_6	UCl_5	UCl_4	UCl_3	UBr_4	UBr_3	UI_4	UI_3
93-Np	NpF_6	NpF_5(?)	NpF_4	NpF_3			$NpCl_4$	$NpCl_3$	$NpBr_4$	$NpBr_3$		NpI_3
94-Pu			PuF_4	PuF_3				$PuCl_3$		$PuBr_3$		PuI_3
95-Am				AmF_3				$AmCl_3$		$AmBr_3$		AmI_3

to prepare only the chloride and bromide of neptunium of oxidation state IV (in addition to the chloride, bromide and iodide of oxidation state III). In the case of uranium, it has been known for some time that there are chlorides of oxidation state higher than IV and a chloride, bromide, and iodide of oxidation state IV. These considerations are well illustrated in Table 2 which lists all of the halides of uranium, neptunium, plutonium, and americium which have been prepared and maintained as stable in the solid state. So far the chemical evidence indicates that it will be difficult, probably impossible, to prepare any of the non-oxygenated halides of americium (and curium) of oxidation state higher than III. In fact, Fried and Florin (*43*) have treated AmF_3 with fluorine at elevated temperatures and have obtained no evidence for the formation of a higher fluoride.

This chemical evidence indicates that the 5*f* electrons are more easily removed by oxidation than the 4*f* electrons, as should be expected on the basis of the predicted lower ionization potentials of 5*f* as compared to 4*f* electrons, provided the hydration and lattice-free energies were not such as to reverse the effect, which might conceivably have been the case. Thus the III state of thorium cannot exist in aqueous solution, and the IV and III states of protactinium are presumably unstable in aqueous solution, although evidence for a valence state lower than V has been reported (*44*, *45*). In the case of solid compounds it has been possible to prepare thorium triiodide (*46*, *47*, *48*) and sesquisulfide (*49*) [as well as lower oxidation states (*47*, *49*)] under rather severe reducing conditions, and it seems likely that tetra- and tripositive compounds of protactinium will be prepared as soon as efforts in this direction are made. In fact, Zachariasen (*50*) and McCullough (*51*) already have some crystallographic evidence for a dioxide of protactinium with the fluorite structure, although this might be a case of solid solution of two oxidation states with a "defect lattice" similar to the solutions studied by Marsh (*52*) and Prandtl and Rieder (*53*) in the PrO_2-Gd_2O_3, TbO_2-Nd_2O_3, and Pr_2O_3-PrO_2 systems and similar to the well-known solid solutions in the iron oxide systems. It is not known whether the unutilized electrons in these lower oxidation states are in the 5*f* or are in the 6*d* configuration, or in some combination of these, since this is just the region where the binding energies for the two types of electrons come closest to equality. From the behavior of uranium, neptunium and plutonium, it must be deduced that as many as three of the 5*f* electrons are given up fairly readily but with increasing difficulty as the atomic number increases.

In this connection it is interesting to note that in the case of the lanthanide elements there are not only several instances of dipositive oxidation states from which 4*f* electrons are lost upon oxidation to the corresponding tripositive states but also in the gaseous atoms there are generally only two electrons (beyond the xenon structure) outside of the 4*f* shell (see Table 5), although the persistent oxidation state is certainly the III state. Another such noteworthy instance is that of cerium sesquioxide, which is so unstable toward oxidation to the dioxide that the former is extremely difficult to prepare and maintain. As Connick (*54*) has pointed out, the stability of the III oxidation state in the rare earth elements is as much a consequence of the hydration and lattice-free energies as of the ionization potentials.

Americium might possess an oxidation state of II which it would attain through the presence of seven electrons in the 5*f* shell in a manner analogous to the II state of europium, the element immediately preceding gadolinium, with

TABLE 3

Oxidation States of Lanthanide and Actinide Elements

Atomic Number	57	58	59	60	61	62	63	64	65
Element	La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb
Oxidation States						2	2		
	3	3	3	3	3	3	3	3	3
		4	4	(4)					4
			(5)						

Atomic Number	89	90	91	92	93	94	95	96
Element	Ac	Th	Pa	U	Np	Pu	Am	Cm
Oxidation States							(2)	
	3	(3)	(3)	3	3	3	3	3
		4	(4)	4	4	4	(4)	
			5	5	5	5	(5)	
				6	6	6		

its seven 4*f* electrons. Because of the greater ease in the removal of the 5*f* electrons, it might require a considerably stronger reducing agent to reduce americium from the III to the II state than is the case for europium. It is not impossible that it will be found that americium can be reduced to the II state in aqueous solution although it is more likely that dipositive solid compounds will be prepared.

Thompson and co-workers (*40*) in some rough preliminary experiments have made partial separations of americium [presumable as Am(II)] from curium in tracer amounts in aqueous solution by using sodium amalgam as the reducing agent and carrying americium selectively with samarium (II) sulfate, and, in addition, by using barium as reducing agent and carrying americium selectively with barium chloride from concentrated hydrochloric acid solution. Curium with its seven 5*f* electrons should exhibit the III state almost exclusively. All of the evidence, obtained from both tracer and macroscopic quantities, indicates that this is the case, but it might not be surprising if it should prove possible to prepare CmO_2 because of the great stability of this dioxide structure (see *Crystallographic Data* on p. 25).

Table 3 summarizes the known oxidation states of the lanthanide and actinide elements in such a way as to bring out the analogy between the two groups and to show the greater ease of oxidation for the members of the latter group. The uncertain or unusual states are designated with parentheses (omitting those states with metallic or possibility of metallic character in their bonding which have oxidation numbers less than III). Such a table has only limited meaning in that oxidation states of solid compounds formed only under drastic conditions, varying in their severity, are listed. However, to list only those states which are stable in aqueous solution also has shortcomings since the existence of an aqueous ion, besides being limited by the ionization potentials of its electrons, is affected by specific chemical processes of hydration and complex formation and is arbitrarily confined within the limits of its oxidation or reduction by water.

The metals of the elements thorium to americium inclusive have been prepared and their properties studied. They bear a striking resemblance to the metals of the rare-earth elements. All of them are highly electropositive, and to about the same degree, which is a similarity to the rare-earth metals and

a difference from the corresponding 5*d* elements, hafnium (element **72**) to iridium (element **77**) inclusive, in which the electropositive character of hafnium is lost as 5*d* electrons are added in going toward iridium. Another remarkable resemblance to the rare-earth elements lies in the densities of the metals. Both americium (*55*) and the analogous 4*f* element, europium (*56*), have densities much lower than their neighboring elements. Thus these metals seem to have radium-like or barium-like structures, with abnormally high radii and analogous electronic structures. A comparison with wolfram, rhenium, osmium, and iridium brings forth no such analogy.

Absorption Spectra in Aqueous Solution and Crystals. One of the characteristic properties of the elements of the lanthanide series, a property which depends upon the 4*f* electrons, is their sharp absorption bands, to a large extent in the visible spectrum. This absorption is due to transitions involving the 4*f* electrons and the sharpness is a consequence of the shielding of these, both in the ground and excited states within the 4*f* shell, by electrons in the outer shells.

The investigations of this type with the elements uranium, neptunium, plutonium, americium, and curium have shown a striking similarity in this property to the rare earths, which is further evidence that we are dealing with 5*f* electrons. The analogy between the rare earths and uranium(IV) in this property was noticed by Goldschmidt (*57*) and between the rare earths and uranium(III) and (IV) by Ephraim and Mezener (*58*) many years ago. This similarity between the actinide and the lanthanide elements is more than qualitative in that the general complexity of the absorption picture undergoes analogous simplification as we approach the middle of the two series, that is, as we approach the elements gadolinium and curium, with their seven 4*f* or 5*f* electrons. Unfortunately, up to the present the most extensive work on the absorption of uranium and the transuranium elements has been done in solution, where much of the sharpness is lost, and with instruments of not very high resolving power.

The aqueous solution absorption spectra of the tripositive actinides, taken from various sources on the Plutonium Project (*59*) are shown in Fig. 1, where the corresponding spectra of the rare-earth elements also are shown for purposes of comparison. Although the absorption curves of Prandtl and Scheiner (*60*) are available for comparison, this work was not done under conditions comparable to those under which the work on the actinides was done. Therefore, Stewart (*61*) has measured the absorption spectra of the rare-earth elements under comparable conditions and his results are also given in Fig. 1. The preliminary results of Lantz and Parker (*62*) on the absorption spectrum of promethium are also included.

It appears that as we approach the middle of the two series (*i.e.*, as we approach the elements gadolinium and curium) the ground states involving the 5*f* electrons uniformly fall increasingly below the next higher states, leading to energy differences for curium of such a magnitude as to cause the absorption to fall outside of the visible in the ultraviolet region as is the case for gadolinium. The analogous peaks of europium and americium at 4000 and 5000 A.U., respectively, have been studied in detail by Jones and Cunningham (*63*), who have shown that in aqueous solution these peaks display striking similarity even in their "fine structure." Sharp absorption bands, undoubtedly involving 5*f* electrons, have also been found in aqueous solution for the other oxidation states of plutonium (*64*) and for the IV and V states of neptunium (*65*).

Such bands are not to be expected and are not found in the case of Np(VI) since this contains only one *5f* electron and hence analogous electronic transitions are not possible.

The best method for the comparison of the absorption spectra of the two groups of elements is to compare the spectra obtained with crystals, where the absorption lines are known to be very sharp for the rare-earth elements on the basis of a large number of measurements with many of these elements. Some such measurements have been made for a number of the transuranium elements and the results so far indicate striking analogies. Freed and Leitz (*66*, *67*) have measured the absorption spectrum of solid americium trichloride and they find sharp lines, of the order of one to five Angstroms wide, which is a width comparable to the sharpest rare-earth spectra. In fact, the sharpness in the spectra (*67*) of americium chloride and americium bromide is so extreme, at room temperature and at 77° K, that only the tripositive europium ion is comparable; since the absorption spectrum of the latter originates from a ground state involving six *4f* electrons, it seems very likely that the basic state of tripositive americium contains six *5f* electrons. They (*67*) also measured the absorption spectra at room temperature and at 77° K of uranium tetrachloride, neptunium tetrachloride, and plutonium trichloride, and the sharpness of the lines indicates that the least stable electrons of these ions are in the inner *5f* shell in the activated as well as in the basic electronic states.

Thus the sharp absorption bands of these elements are to be ascribed to

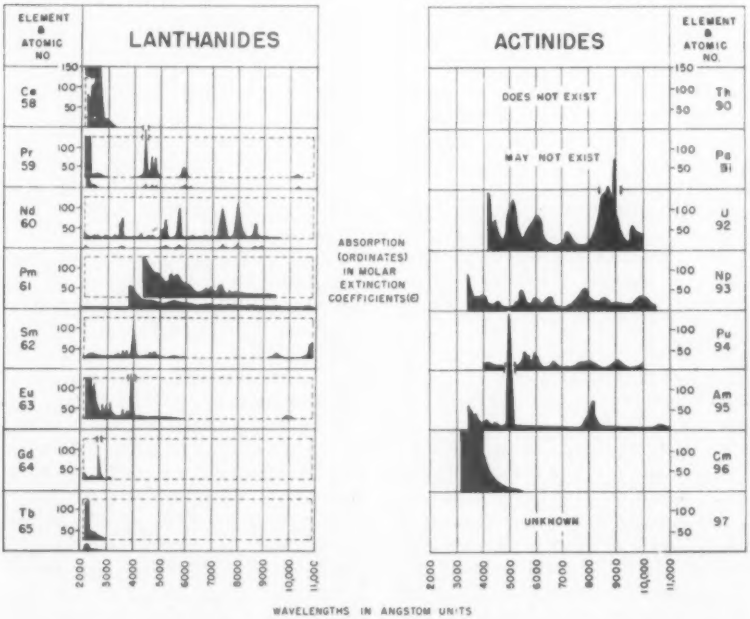

FIG. 1. Absorption spectra of aqueous tripositive lanthanides and actinides

electronic transitions within the 5*f* shell, in which the $5f^n$ configuration is preserved in the upper and lower states for a particular ion.

Crystallographic Data. As mentioned above, crystallographic evidence in addition points to the filling of the 5*f* shell in this neighborhood of heavy elements. Some years ago, Goldschmidt (*68, 20, 69*) had already noticed the isomorphism of ThO_2 and UO_2, and the decrease in size in going to UO_2, and had interpreted this to indicate the presence of 5*f* electrons in uranium. The observation of Zachariasen (*50*) of the isomorphism of the compounds ThO_2, PaO_2, UO_2, NpO_2, PuO_2, and AmO_2 (*70*), together with his observation of the regular decrease in radius of the metallic ion in these oxides, has been interpreted by him to be excellent evidence that the electrons are going into the 5*f* shell. In this series, however, the lattice constants of the PaO_2 (*50, 51*), possibly also AmO_2 (*71*), do not fit perfectly into the regular pattern for decrease in atomic radius of the metallic ions; this might be due to mixed oxidation states with defect structures (*72*) similar to the situation found for the higher praseodymium oxides (*52*) and for the oxides of uranium (*73*) and neptunium (*74*). The relative ease with which PaO_2 and AmO_2 may be formed, just by heating in air, which is in sharp contrast to the instability of the tetrapositive states of these elements in aqueous solution, illustrates the great stabilizing influence of the dioxide crystal lattice. Both of these investigators have advanced the hypothesis that it is a "thoride" series, *i.e.*, that the first 5*f* electron appears in the first element beyond thorium, namely protactinium.

Zachariasen has used the X-ray diffraction method to determine the molecular structures of a great number of compounds of thorium, uranium, and the transuranium elements. All of these measurements point toward the filling of the 5*f* shell since analogous compounds are found to be isomorphous, indicating that the successive electrons are added in such a way (*i.e.*, to an inner shell) as to allow the analogous compounds of successive elements to have identical molecular structures. Thus, Zachariasen (*75*) finds that practically all of the various halide types shown in Table 2 have isomorphous structures; for example, all members of the group ThF_4-UF_4-NpF_4-PuF_4 are of identical structure type, all members of the group UF_3-NpF_3-PuF_3-AmF_3 are also isomorphous with each other, and the same is true for the group UCl_3-$NpCl_3$-$PuCl_3$-$AmCl_3$, *etc.* To be sure, in some cases (for example, UBr_3-$NpBr_3$-$PuBr_3$-$AmBr_3$), there is a change in structure type in proceeding up the group, but this is to be expected on the basis of the contraction which takes place, and is entirely consistent with the addition of the successive electrons to the 5*f* shell.

Zachariasen (*75*) has used these structure data to calculate ionic radii, and these radii show a progressive decrease in size with increasing atomic number in a manner quite analogous to the well-known "lanthanide contraction" observed with the rare-earth elements, the compounds of which are in turn isomorphous with the corresponding compounds of the actinide elements. To illustrate these considerations further, Table 4 gives the ionic radii of a number of the actinide and lanthanide elements (*75*) with interpolated values given in parentheses.

Magnetic Susceptibility Data. Magnetic susceptibility measurements on compounds of the heaviest elements ideally should lead to the resultant magnetic moments in fundamental units and in this way give information on the quantum states of the responsible electrons. Actually, as evidenced by the rare-earth elements, the situation is

TABLE 4

Ionic Radii of Actinide and Lanthanide Elements

No. of 4f or 5f Electrons	*Actinide Series*				*Lanthanide Series*	
	III *State*		IV *State*			
0	Ac^{+3}	1.11 A.U.	Th^{+4}	0.95 A.U.	La^{+3}	1.04 A.U.
1	(Th^{+3})	(1.08)	Pa^{+4}	0.91	Ce^{+3}	1.02
2	(Pa^{+3})	(1.06)	U^{+4}	0.89	Pr^{+3}	1.00
3	U^{+3}	1.04	Np^{+4}	0.88	Nd^{+3}	0.99
4	Np^{+3}	1.02	Pu^{+4}	0.86	Pm^{+3}	(0.98)
5	Pu^{+3}	1.01	Am^{+4}	0.85	Sm^{+3}	0.97
6	Am^{+3}	1.00			Eu^{+3}	0.97

rather complex and the exact behavior expected for the heaviest elements, on the basis of the presence of either 5*f* or 6*d* electrons, cannot be, or at least has not been, predicted. Nevertheless, such measurements should give, and have given, information on this point.

The earliest magnetic susceptibility measurements that were made on the compounds of uranium (*76*, *77*, *78*, *79*) and plutonium (*80*) showed that these are paramagnetic, yet the results are difficult to interpret quantitatively. A simple qualitative explanation of the magnetic susceptibilities of Pu(III), Pu(IV) and Pu(VI) lies in the assumption that there are five 5*f* electrons in Pu(III) which are successively removed as one goes to the higher oxidation states. These measurements, however, do not lead to this interpretation as the sole and unambiguous one, and as a result must be regarded only as being consistent with and lending weight to this view, but not giving proof of it.

Later Hutchison and Elliott made magnetic susceptibility measurements over a wide range of temperatures on a number of solid compounds of the heaviest elements. In the case of uranium(IV) compounds (*81*, *82*) they found that a number behave in a manner similar to praseodymium(III) compounds, indicating that these two groups of compounds have isoelectronic structures with respect to *f* electrons and thus have two such electrons. They disclosed, in addition, that the temperature dependence of the magnetic susceptibility of these uranium(IV) compounds obeys the Curie-Weiss law over a range of temperatures, and through extrapolation with the use of this law they deduced a resultant magnetic moment very close to that expected for two *f* electrons. They also concluded that the crystal fields produce more pronounced perturbing effects in this case than in the corresponding case involving 4*f* electrons. Their measurements (*82*) on neptunium(V), which is isoelectronic with uranium(IV), also indicated the presence of two *f* electrons here.

Howland and Calvin (*83*) have measured the magnetic susceptibilities of the cations of uranium, neptunium, plutonium, and americium in most of their stable oxidation states in aqueous solution. To account consistently for the observed values of the magnetic susceptibilities, the heavy atoms must be assumed to have electronic configurations (beyond the radon structure) of the type $(5f)^{1-6}$. For example, Np(VI) corresponds to the structure $5f$; U(IV), Np(V), and Pu(VI) to $5f^2$; Np(IV) to $5f^3$; Pu(IV) to $5f^4$; Pu(III) to $5f^5$; and Am(III) to $5f^6$.

The experimental effective magnetic

moments are generally lower than the theoretical values and lower than experimental values for corresponding lanthanide $4f^n$ cations, as would be expected on the basis of the Stark effect produced by electric fields of anions and of water dipoles. Failure of the Russell-Saunders approximation to the coupling between electrons may account for some of the error in the theoretical calculations. That the susceptibilities of Pu(III) and Am(III) are many-fold lower than those of Sm(III) and Eu(III), respectively, is attributed to wider multiplet splitting in the actinide atoms. Figure 2 shows a plot comparing the experimental magnetic susceptibilities of the lanthanide and actinide cations in such a way as to show their remarkable analogy in this property.

Spectroscopic Data. Spectroscopic evidence also lends support to the actinide interpretation. Kiess, Humphreys, and Laun (*84*, *85*) investigated the spectrum of uranium atoms and gave the interpretation that the electron configuration of the lowest state of neutral uranium is $5f^36d7s^2$ (beyond radon), a configuration which fits in well since uranium is the third element in the series. Other work by Schuurmans and co-workers (*86*, *87*) on the spectrum of gaseous U^+ and U and by McNally and Harrison (*88*) on U^+ has given results which are consistent with this as the ground term for the neutral uranium atom. Other observations (*89*, *90*, *91*, *92*) made on the gaseous Th^+ spectrum indicate that the *5f* is very close to the *6d* electron in binding energy in the neutral, free thorium atom. Russell (*93*) has made a complete analysis of the X-ray data for radium, thorium, and uranium and has concluded that the *5f* lies lower than the *6d* level and that the *5f* shell begins to fill at thorium.

Tompkins and Fred (*94*) have made a qualitative comparison of the emission spectra of the actinide and the lanthanide elements. They found such a strong analogy between the average intensity of the lines in the case of americium and europium that it seems safe to conclude that these have similar electronic structures in the ground states. This then indicates the configuration $5f^77s^2$ for the gaseous atom of americium.

TABLE 5

Suggested Electron Configurations (Beyond Radon and Xenon) for Gaseous Atoms of Actinide and Lanthanide Elements

89-Ac	$6d7s^2$	57-La	$5d6s^2$
90-Th	$6d^27s^2$ (or $5f6d7s^2$)	58-Ce	$4f^26s^2$
91-Pa	$5f^26d7s^2$ (or $5f6d^27s^2$)	59-Pr	$4f^36s^2$
92-U	$5f^36d7s^2$	60-Nd	$4f^46s^2$
93-Np	$5f^57s^2$ (or $5f^46d7s^2$)	61-Pm	$4f^56s^2$
94-Pu	$5f^67s^2$ (or $5f^56d7s^2$)	62-Sm	$4f^66s^2$
95-Am	$5f^77s^2$	63-Eu	$4f^76s^2$
96-Cm	$5f^76d7s^2$	64-Gd	$4f^75d6s^2$

Correlations and Deductions

Electronic Configurations. Table 5 gives what appears to be the configuration or the best prediction for the configuration (beyond the radon structure)

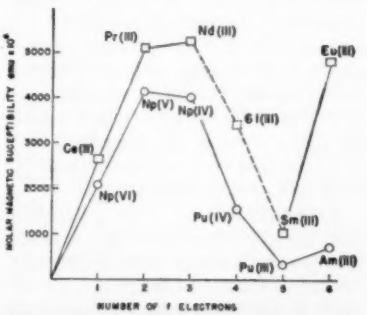

FIG. 2. Plot of molar magnetic susceptibilities of some aqueous actinide and lanthanide ions

of the ground state of the neutral gaseous atom for each of the elements actinium to curium inclusive. The trend in the chemical properties, with its implication that the 5*f* becomes progressively of lower energy compared to the 6*d* level as the atomic number increases, has been used as an aid in making the predictions.

The configurations (*95*, *96*, *97*) (beyond xenon) of the corresponding neutral rare-earth elements are given for comparison. The ground states given for cerium and praseodymium are those predicted (*95*, *97*) on the basis of the recently determined ground states of the singly ionized atoms (*98*), and that of promethium is obtained by interpolation, and as a consequence these are subject to some doubt. The ground states given for neodymium (*86*), samarium, europium and gadolinium are those spectroscopically determined (*98*) for the neutral atoms and should be considered as well established.

It should be emphasized that it would be entirely consistent from the point of view that we are dealing here with a series of actinide elements if it should eventually be found that there are no 5*f* electrons present in thorium (or protactinium). It is quite possible on the basis of present evidence that protactinium, or even uranium, might be the first to have 5*f* electrons. It seems quite likely, however, that electrons will be placed in the 5*f* shell earlier in the series than uranium and that neutral protactinium will have at least one such. An essential point is that curium definitely seems to have seven 5*f* electrons and element 103 probably would have fourteen 5*f* electrons.

In the case of some of the elements in the series it may be difficult to assign electrons to the 5*f* or 6*d* shells since the energy necessary for the shift from one shell to the other may be within the range of chemical binding energies. Much of the experimental data already described, such as the ease of oxidation to higher states and the more complicated situation for the magnetic susceptibilities as compared to the rare-earth elements, points to lower binding and less electrostatic shielding by outer electrons of 5*f* compared to 4*f* electrons, just as should be expected, and the magnitude of these effects seems reasonable.

The electronic configuration may differ from compound to compound or even with the physical state of a given compound. Moreover one certainly cannot be sure that the configuration of the gaseous atom, for example, will correspond to that of the compounds or that of the hydrated ions in solution. In the case of the lanthanides, in fact, the configuration of the gaseous atom has, in general, only two electrons (beyond the xenon structure) outside of the 4*f* shell, although the predominant oxidation state is certainly the III state. Since the energy difference between such far outlying levels as the 5*f* and 6*d* shells is rather small and since resonance effects should be rather large, these may predominate in determining that a composite energy level lies lowest. Thus some of these elements could possibly constitute what might more properly be called a 5*f*–6*d* range in this series, rather than a part of a totally 5*f* transition group.

The evidence that has accumulated so far seems nevertheless to point to lower energies, for the 5*f* compared to the 6*d* levels for the compounds of the element, as early as uranium in this series. It is in the case of the elements thorium and protactinium where the relative energy position of these levels is as yet most uncertain. As in the other transition series, the relative energy position of the shell which is undergoing the filling process becomes lower as the successive electrons are added, and by the time americium and curium, and presumably the subsequent

members of the series, are reached, the 5*f* seems clearly to be definitely of lower energy than the 6*d* shell. The electronic structures proposed for neptunium and plutonium are influenced by the possibility of analogy with their homologous rare-earth elements, but since it is not yet possible to place these electrons with confidence, alternative structures for gaseous neptunium and plutonium are suggested in Table 5. It is reasonable to expect the ground states of the gaseous elements beyond uranium in this series to maintain no electrons in the 6*d* shell except in the cases of those elements with the half-filled and filled 5*f* shell (curium and element 103).

It seems quite possible for those elements where the energies are so nearly equal to have the 5*f* as well as the 6*d* (and outer orbitals) involved in the chemical binding in some compounds and complex ions, an interesting possibility for a new type of bonding. Such bonding involving 5*f* orbitals should be the subject for interesting future experimentation.

Figure 3 purports to show, in an extremely rough and qualitative way, a pictorial representation of the binding energy of the most easily removable 5*f* and 6*d* electron (of those present) for each of the heaviest elements. A rough representation such as this can be justified only if it helps somewhat in the understanding of the situation, and it is hoped that this is the case here.

Possible Deductions Without Data on Transuranium Elements. Although it is the information on the transuranium elements that has been decisive in enabling us to come to the present view concerning the electronic structure, or more properly speaking, the best position in the periodic table for the heaviest elements, it is interesting to conjecture, in retrospect, about the possibility of having arrived at a similar conclusion without this information. Actually there has been much information about

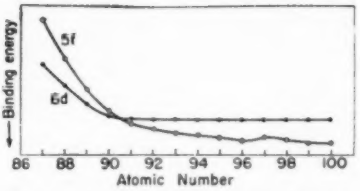

FIG. 3. Qualitative representation of electronic binding energies in the heaviest elements

actinium, thorium, protactinium, and uranium, especially about the latter, which pointed in this direction.

As mentioned above, there is the similarity among the metals of these elements with respect to electropositive character. In addition, the melting point of uranium metal seems to relate it more to the immediately preceding elements than to wolfram and molybdenum. The analogy of uranium ions to the rare-earth ions with respect to light absorption and the spectroscopic evidence for a ground state of the gaseous uranium atom involving three 5*f* electrons has already been mentioned.

Uranium differs considerably from wolfram (and molybdenum) in the chemistry of the lower oxidation states and, in fact, uranium(III) has great similarity to the tripositive rare-earth elements and actinium, while uranium(IV) resembles thorium and cerium(IV). Thus uranium(III) and uranium(IV) are not acidic in character, do not tend like wolfram and molybdenum to form such exceedingly strong complex ions in solution, have fluorides which are insoluble and isomorphous with the fluorides of the rare-earth elements and have other halides with crystal structures which are in general isomorphous with the corresponding rare-earth halides. On the other hand, wolfram(III) and wolfram(IV) exist in aqueous solution predominantly as strong complex ions; for example, wolfram(III) has a strong chloride complex ion and wolfram(IV) forms strong fluoride and

cyanide complex ions. In this connection Thompson (*99*) has pointed out that wolfram(IV) forms the very stable complex ion $W(CN)_8^{-4}$, with the stable configuration of eighteen outer electrons, while uranium(IV) possesses no significant tendency to form an analogous complex cyanide ion as would be expected if uranium possessed the same outer electronic structure as wolfram. In the VI oxidation state there is the distinctive uranyl ion (UO_2^{++}), with no analogues in wolfram or molybdenum, and compounds such as the diuranates in contrast to the soluble wolframates and molybdates.

Although molybdenum dioxide and wolfram dioxide have isomorphous crystal structures (rutile type), wolfram dioxide and uranium dioxide do not; uranium dioxide, thorium dioxide, and cerium dioxide, however, all possess isomorphous (fluorite) structures. Thorium often resembles Ce(IV) more than it does hafnium and zirconium as, for example, in the insolubility of thorium and cerium(IV) fluorides in contrast to the solubility of the fluoride of hafnium and zirconium.

It is interesting to note that although uranium is not associated with wolfram in minerals, uranium and thorium minerals practically always have the rare-earth elements associated with them and the rare-earth minerals practically always contain uranium or thorium.

Arguments on the basis of the evidence from the chemical properties of thorium and uranium have been given by others, including Villar (*19*), more recently Stedman (*100*), and very recently Szabo (*101*) for a 5*f*-type transition series in the heaviest elements, beginning with thorium.

1 H 1.008									
3 Li 6.940	4 Be 9.02								
11 Na 22.997	12 Mg 24.32								
19 K 39.096	20 Ca 40.08	21 Sc 45.10	22 Ti 47.90	23 V 50.95	24 Cr 52.01	25 Mn 54.93	26 Fe 55.85	27 Co 58.94	28 Ni 58.69
37 Rb 85.48	38 Sr 87.63	39 Y 88.92	40 Zr 91.22	41 Nb 92.91	42 Mo 95.95	43 Tc	44 Ru 101.7	45 Rh 102.91	46 Pd 106.7
55 Cs 132.91	56 Ba 137.36	57 La 138.92	58-71 SEE La SERIES	72 Hf 178.6	73 Ta 180.88	74 W 183.92	75 Re 186.31	76 Os 190.2	77 Ir 193.1	78 Pt 195.23
87 Fr	88 Ra	89 Ac	SEE Ac SERIES						

FIG. 4. Periodic table showing heavy elements as members

LANTHANIDE SERIES	57 La 138.92	58 Ce 140.13	59 Pr 140.92	60 Nd 144.27	61 Pm	62 Sm 150.43	63 Eu 152.0	64 Gd 156.9
ACTINIDE SERIES	89 Ac	90 Th 232.12	91 Pa 231	92 U 238.07	93 Np 237	94 Pu	95 Am	96 Cm

Position in Periodic Table and Nomenclature. Soon after the establishment of the concept of atomic number, the rare-earth elements were properly fitted into the classification of the elements and the periodic table took its present form, in which there is general agreement as to the various groups and subgroups, with differences only in regard to the best geometrical arrangements for presenting the information. Thus even the undiscovered elements with atomic numbers within the confines of this classification had their places fixed, and upon their discovery no reasons were found to change their positions. However, this is not the situation with respect to the transuranium elements. Their positions could be fixed only after experimental determination of their properties and apparently even influence the positions of elements which had been previously given places.

Since this situation presents a new problem, it is necessary to consider carefully the basis which has given rise to the present classification, in order to try to find the traditional criteria to apply to the elements now under consideration. The chemical properties, especially in aqueous solution, have been an important criterion. Thus the rare-earth elements assumed a position in recognition of their predominant tripositive character in aqueous solution, a property which is not deducible from the electronic structure of the atomic ground states. However, the spectroscopic data have determined the general regions of the transition series (where the $3d$, $4d$, $5d$, and $4f$ shells undergo filling) and seem to have influenced the practice of uninterrupted

							1 H 1.008	2 HE 4.003
			5 B 10.82	6 C 12.010	7 N 14.008	8 O 16.000	9 F 19.00	10 Ne 20.183
			13 Al 26.97	14 Si 28.06	15 P 30.98	16 S 32.06	17 Cl 35.457	18 A 39.944
28 Ni 58.69	29 Cu 63.57	30 Zn 65.38	31 Go 69.72	32 Ge 72.60	33 As 74.91	34 Se 78.96	35 Br 79.916	36 Kr 83.7
46 Pd 106.7	47 Ag 107.880	48 Cd 112.41	49 In 114.76	50 Sn 118.70	51 Sb 121.76	52 Te 127.61	53 I 126.92	54 Xe 131.3
78 Pt 195.23	79 Au 197.2	80 Hg 200.61	81 Tl 204.39	82 Pb 207.21	83 Bi 209.00	84 Po	85 At	86 Rn

64 Gd 156.9	65 Tb 159.2	66 Dy 162.46	67 Ho 163.5	68 Er 167.2	69 Tm 169.4	70 Yb 173.04	71 Lu 174.99

96 Cm							

placing of the transition elements in subgroups (that is, all ten 3*d* transition elements from $_{21}$Sc to $_{30}$Zn are usually uninterruptedly placed in subgroups, and the same for the 4*d*, 5*d* and 4*f* transition series).

Using such criteria, the best method of presenting the actinide elements in the periodic table seems to be that shown in Fig. 4. Here are shown the fourteen elements of atomic numbers 90 to 103 inclusive, with actinium (element 89) as the prototype listed as a series below, and in a manner similar to, the common listing of the fourteen rare-earth elements of atomic numbers 58 to 71 inclusive, for which lanthanum (element 57) is the prototype.

It is not proposed that this particular form of the periodic table has any more merit than any of a number of others which place these elements in positions homologous to the rare-earth elements, since it is obvious that they can be analogously placed in a number of other types of tables or charts. The elements 90 to 96 inclusive or the first few of them could in addition be listed separately below the 5*d* elements in recognition of the resemblance of the first few of these to 5*d* elements. This appears to be undesirable, however, since the last members of this group bear no such resemblance and it is probably impossible to draw a line as to just where the resemblance ends.

Following the suggestion (*36*) that the information on the transuranium elements appeared to have reached such a state as to make it possible definitely to place the heaviest elements in the periodic table (as an actinide transition series), a number of publications (*102*, *103*, *104*, *105*, *106*, *107*, *108*, *109*, *110*) commenting on this proposal have appeared. These papers are in general agreement with the suggestion, although in some cases the reasons therefor differ and in others there is an understandable expression of desire to see more of the evidence in detail. A couple of publications (*111*, *112*) indicate a preference for the placement of the transuranium elements in the periodic table as a uranide group in spite of the evidence which has been presented concerning their chemical properties. A number of variations of periodic tables in which these elements have been placed as actinides homologous to the lanthanides have been published recently (*113*, *114*, *115*, *116*, *117*, *118*, *119*, *120*, *121*).

As mentioned above, a very important point is the presence of seven 5*f* electrons in stable, tripositive curium (element 96), making this element very actinium-like. A series of "thoride" elements, for example, would imply stable IV oxidation states in elements 95 and 96, with the presence of seven 5*f* electrons and the IV state almost exclusively in element 97. A series of this type seems to be ruled out by the now known instability of americium in solution in the IV state and by the apparent nonexistence in aqueous solution of any oxidation state other than III in curium. Moreover, the III state of uranium would be surprising on this basis since this element would be the *second* member of a "thoride" or "IV oxidation state" series. The fact that nearly a year was spent in an unsuccessful effort to separate tracer amounts of americium and curium from the rare earths, immediately following the discovery of the former, illustrates how unnatural it would be to regard them as members of a "thoride" or "IV oxidation state" group.

The group probably could have been just as well described by some other term such as "curide series," rather than "actinide," which is derived from straight analogy with the term lanthanide. Another possibility would be to use a term such as "type 5*f* rare earths" or another name analogous to "rare earths." A suggestion here

might be "synthetic earths" in view of the synthetic source of all except the first three members. (The best source of actinium is synthetic, coming from pile neutrons by the reactions $Ra^{226}(n,\gamma)Ra^{227} \xrightarrow{\beta^-} Ac^{227}$.) Irrespective of the name which usage will finally assign to this group of elements, however, it seems that the outstanding characteristics of the group, namely the "eka-gadolinium" character of curium (and the presumed "eka-lutetium" character of element 103), together with the regularly increasing trend toward actinium-like character in going from thorium to curium, are best represented by listing these elements in corresponding positions under the rare-earth elements if it is desirable to give each element only one place in the table.

Predicted Properties of Transcurium Elements. There has been a good deal of speculation concerning the upper limit of atomic number for the existence of elements, with considerations that such a limit might arise from either atomic or nuclear (radioactivity or spontaneous fission) instability. If the former is not the limiting factor, it appears likely, on the basis of extrapolation from the nuclear properties of the heaviest elements, that at least the first few elements above element 96 will have isotopes of sufficiently long life to make possible their investigation at least on the tracer scale should the problem of their production in detectable amount be solved.

It is interesting to speculate about the chemical properties of these undiscovered elements beyond curium. The seven elements immediately following, that is, elements 97 to 103 inclusive, should constitute the second half of this rare-earth-like transition group. It appears likely that the added electrons in proceeding up this series will be placed in a 5*f* shell of definitely lower energy than the 6*d* shell. Element 97 will probably have a IV as well as III oxidation state, and, in view of the lower binding energy of the 5*f* as compared to the 4*f* electrons, it might be easier to oxidize element 97 (eka-terbium) to this IV state than it is in the case of terbium, since the hydration and lattice energies do not appear to reverse the simple predictions for oxidation potentials from ionization potentials in these elements. Correspondingly, it should be easier to oxidize element 98 (eka-dysprosium) to the IV and V oxidation states than it is in the case of dysprosium, for which, in fact, oxidation above the III state is practically impossible. Toward the end of the series, elements 102 and 101 should be capable of being reduced to the II oxidation state, analogously to ytterbium and thulium, while element 103 should be analogous to lutetium with respect to the complete stability of the III state.

Element 104 should continue with the filling of the 6*d* shell and be a true eka-hafnium. After the filling of the 6*d* shell in the following elements, there would be addition to the 7*p* shell with the attainment of the rare gas structure at hypothetical element 118 (on the logical assumption that the 5*g* shell does not start to fill before this point).

Summary

All of the available evidence leads to the view that the 5*f* electron shell is being filled in the heaviest elements, giving rise to a transition series which begins formally with actinium in the same sense that the rare-earth or "lanthanide" series begins with lanthanum. Such an "actinide" series is suggested on the basis of evidence in the following lines: (1) chemical properties, (2) absorption spectra in aqueous solution and crystals, (3) crystallographic structure data, (4) magnetic susceptibility data, and (5) spectroscopic data. This series differs from the rare-earth series in having more

oxidation states above the III state and in other ways, all connected with the lower binding of 5*f* compared to 4*f* electrons. The salient point is that the characteristic oxidation state (*i.e.*, the oxidation state exhibited by the member containing seven 5*f* and presumably also by the member containing fourteen 5*f* electrons, curium and element 103) is the III state, and the group is placed in the periodic table on this basis. The data also make it possible to give a suggested table of electronic configurations of the ground state of the gaseous atom for each of the elements from actinium to curium inclusive.

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Calorimetric Radioactivity Measurements

Where radioactivity levels of millicuries or curies must be determined, microcalorimetry obviates many of the uncertainties of the counting method. The author here reviews the development of the instruments and techniques used in this field, and discusses basic present-day practices

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PERHAPS THE MOST EXACT method for determining radioactivity is to measure the rate of heat evolution by means of a microcalorimeter. The calorimetric method requires a larger sample activity than does the counting method, but it is a useful procedure where activity levels of the order of millicuries or curies are to be determined. It involves few of the uncertainties of the counting method.

Alpha disintegration rates and half-lives may be determined readily with an accuracy of one percent if the energy of the disintegration and the isotopic mass are known. To illustrate such a determination we shall consider the work of Stout and Jones (*60*) on Pu^{239}, mass 239.08. They observed a heating effect of 4.59×10^{-4} cal gm^{-1} sec^{-1} or 7.63×10^{-18} erg sec^{-1} per atom. From a measurement of the alpha particle range and a consideration of the recoil energy, the disintegration energy was taken as 5.23 Mev or 8.38×10^{-6} ergs. The half-life of Pu^{239} was computed as

$$T_{1/2} = \frac{\ln 2}{\lambda} = \ln 2 \frac{N}{dN/dt}$$

$$= \frac{0.69315 \times 8.38 \times 10^{-6}}{7.63 \times 10^{-18}} \text{ sec}$$

$$= 7.59 \times 10^{11} \text{ sec or } 2.41 \times 10^{4} \text{ y}$$

Calorimetric determinations of mean beta energies have been of great theoretical importance as a verification of values obtained by integration of the observed beta-ray spectra. Here we may compare the results for Bi^{210} (RaE):

$\bar{E}_{\beta(calor.)} = 0.344 \pm 0.034$ Mev
[Ellis and Wooster (*19*)]
$= 0.337 \pm 0.020$ Mev
[Meitner and Orthmann (*29*)]
$= 0.320 \pm 0.005$ Mev
[Zlotowski (*74*)]
$\bar{E}_{\beta(spect.)} = 0.340$ Mev
[Neary (*76*)]

and for tritium:

$\bar{E}_{\beta(calor.)} = 5.69$ kev
[Jenks, Ghormley, and Sweeton (*23*)]
$\bar{E}_{\beta(spect.)} = 5.5$ kev
[Curran, Angus, and Cockroft (*77*)]

The use of a calorimeter in a determination of $\bar{E}_\beta$ is analogous to the use of a primary standard in quantitative analysis. It lacks general applicability because an accurate knowledge of the absolute disintegration rate of the sample is required. This knowledge is usually difficult to obtain. On the other hand, if $\bar{E}_\beta$ is known from the tested spectrographic integration method, it is possible to obtain quite accurate values of absolute beta disintegration rates.

For the measurement of gamma-ray energies and intensities it is necessary to have a large mass of absorber in order to reduce the uncertainty regarding the efficiency of absorption. In general,

Useful Conversion Factors in Calorimetry*

1 rutherford (rd)†	= 10^6 disintegrations per second
1 curie	= 3.7×10^{10} disintegrations per second‡
1 calorie (cal_{15})	= 4.1847 international joules
	= 4.1855×10^7 ergs
	= 2.6126×10^{13} (energy of one absolute Mev)
1 cal_{15} per hour	= $(7.2573 \times 10^3)/\overline{E}$ rd
	= $0.19614/\overline{E}$ curie
1 watt	= 1.0002×10^7 ergs per second
	= 6.2433×10^{12} (energy of one absolute Mev) per second
	= $(6.2433 \times 10^7)/\overline{E}$ rd
	= $(1.6874 \times 10^3)/\overline{E}$ curie

* Except as noted these formulas are based on values reported by R. T. Birge, *Revs. Mod. Phys.* **13**, 233 (1941). $\overline{E}$ is the average disintegration energy in millions of absolute electron volts.

† The rutherford is the absolute unit recommended by the U. S. National Bureau of Standards.

‡ This is a fixed value recommended by the International Radium Standard Commission.

the calorimeters which have been used thus far are unable to cope with the inherent high heat capacity of such an absorber. (A possible solution to this difficulty might be to operate the calorimeter at very low temperatures to reduce the heat capacity of the calorimeter.) As a natural consequence almost no work has been reported concerning heat effects caused by gamma radiation. This is true in spite of the present-day need in nuclear physics for a direct integration method to obtain gamma-ray energies.

Differential and Twin Microcalorimeters

The first use of calorimeters in radioactivity measurements was in 1903, when Pierre Curie and A. Laborde (*8*) undertook to verify the belief, not altogether obvious at that time, that the heat produced by a radioactive substance is due to absorption of its energetic radiations. That investigation might be considered the beginning of microcalorimetry also, because a need was created for precise measurement of very small quantities of heat.

Curie and Laborde used two matched Dewar flasks in their first investigation. One contained a gram of a mixture of about one-sixth by weight of radium bromide in barium bromide. A similar sample of one gram of barium bromide was placed in the other. With this equipment they were able to estimate that about 100 calories were released by one gram of radium bromide in one hour.§

This result was confirmed by means of a small Bunsen ice-calorimeter. In the next year, Curie and Dewar (*7*) used liquid air and liquid hydrogen in separate proofs that the heat developed by a radioactive substance is independent of the temperature.

This method for the determination of small amounts of heat, known as *twin differential* microcalorimetry, has been considerably improved and remains the most satisfactory system in many applications. The first improvements were provided by K. Ångström (*1*) in 1905, Schweidler and Hess (*42*, *43*) in 1908.

§ A statement made by Curie and Laborde (*8*) as a result of this work might be of historical interest: "Si l'on cherche l'origine de la production de chaleur dans une transformation interne, cette transformation doit être de nature plus profonde et doit être due à une modification de l'atome de radium lui-même."

Pegram and Webb (*31*) in 1908, and Callendar (*5*) and Poole (*32*) in 1910.

Calorimeters became adequately insulated and were placed in thermostats; use was made of a compensating electric heater in the second calorimeter of a pair. The heater current was evaluated when the rate of heating of both calorimeters was identical, as measured by thermocouple joints attached to each. With slight modifications the *compensation current* method has been used in more recent times by Wertenstein (*66*), Yovanovitch (*70*), and Dorabialska (*9*, *10*).

In 1913 Rutherford and Robinson (*39*) used small platinum coils of about 100 ohms resistance, attached to the inside of duplicate glass tubes which were in turn placed in brass tubes in a water thermostat. The coils served as arms of a Wheatstone bridge which was completed, as shown in Fig. 1, by two 100-ohm manganin coils kept at constant temperature. The calorimeter could be used as a null instrument as in the *compensation current* method, or it could be calibrated against known heating rates produced by the electric heater. Rutherford found that one gram of radium in equilibrium with its short-life products produced 134.7 cal/hr.

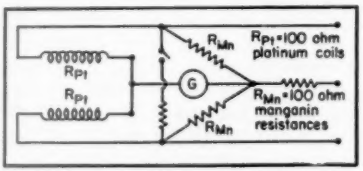

FIG. 1. Rutherford-Robinson resistance thermometer circuit

Earlier, in 1903, Rutherford and Barnes (*37*, *38*) described a differential air twin calorimeter in which the value of the heat emitted by a gram of radium in equilibrium with its daughters was estimated to be 110 cal/hr. In this device, diagrammed in Fig. 2, no provision was made for complete absorption of beta and gamma radiation, and the low value obtained is not indicative of the precision of the method. Differences in the height of the xylene column were observed with a telescope.

Duane (*16*) applied somewhat similar principles to an ether differential calorimeter. The two calorimeter flasks were protected from heat exchange with the surroundings by cotton insulation. According to Duane, the sensitivity is great enough to measure the amount of heat produced in one hour to within 0.001 cal. Unequal distribution of temperature at the ether surface is probable in the design shown in Fig. 2, and it would have limited accuracy.

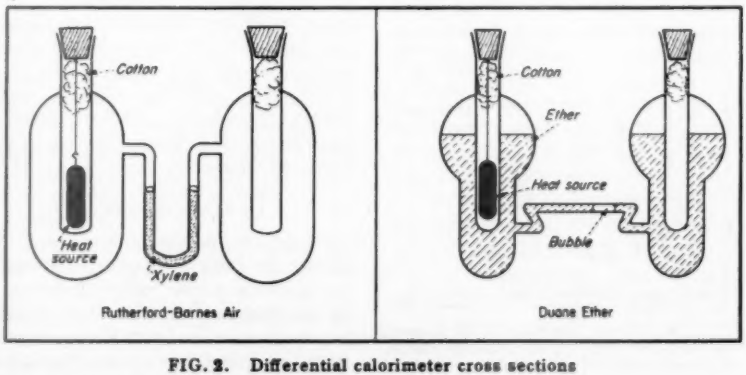

FIG. 2. Differential calorimeter cross sections

Dorabialska (*11*) has also reported results with this type of calorimeter.

Ellis and Wooster (*17*, *18*) used a specially designed differential calorimeter to measure the heating effect of only the gamma radiation of RaB and RaC in equilibrium. Heat due to alpha and beta radiation was balanced by transferring it equally to both metallic segments of the quadrant calorimeter employed (Fig. 3).

The active radon source in a sealed glass tube was placed in the hollow axis of a copper rod. Two opposite quadrants of the calorimeter were of balsa wood; the third was lead; and the fourth

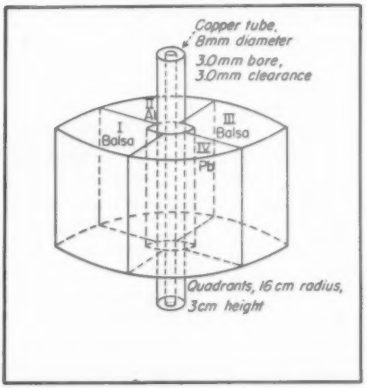

FIG. 3. Ellis and Wooster differential gamma calorimeter

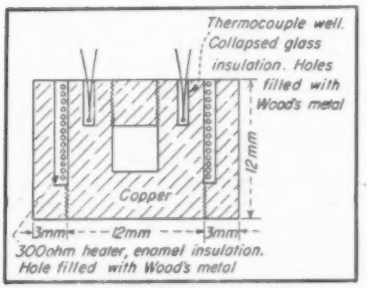

FIG. 4. Cross section of Orthmann microcalorimeter. Two of these assemblies are used for differential calorimetry

was aluminum, bored out to present the same heat capacity as the lead segment. Temperature differences, caused by differential gamma-ray absorption, were measured by a thermopile. The system was surrounded by balsa wood and a copper sheath. From a determination of the temperature difference and the relative absorption coefficients in aluminum and lead, they reported the heating effect of the total gamma radiation by RaB and RaC in equilibrium with one gram of radium to be 8.6 cal/hr; 0.86 cal/hr due to RaB and 7.7 cal/hr due to RaC.

Orthmann (*30*) has presented details of construction of a differential calorimeter for absolute determination of as little as 0.005 cal/hr with a probable error of not more than 0.0002 cal/hr. It consisted of two identical copper cylinders 12 mm in diameter and 12 mm high, with a hole in each, as shown in Fig. 4, to receive a radioactive sample. A 300-ohm heater coil on each provided for calibration, and a 15-element thermopile was used to detect temperature differences. Each calorimeter weighed about 11.4 gm and had a heat capacity of 1.0 cal/degree C. The system was evacuated to 10^{-6} mm Hg. It was kept in a constant temperature bath.

Among the other differential microcalorimeters which have been employed in radioactivity determinations are those of Ellis and Wooster (*19*), Lange and Messner (*25*), Rump (*36*), Steiner and Johnston (*46*), Bayne-Jones (*3*), Ward (*65*), Gucker, Pickard, and Planck (*20*), Magee, DeWitt, Smith, and Daniels (*27*), Mason (*28*), Tonnelat (*64*), and Greinacher (*21*).

Ice Microcalorimeters

Precht (*33*, *34*, *35*) was the first to modify the Bunsen ice calorimeter into an instrument for microcalorimetry. In the Bunsen calorimeter the change in volume associated with the melting of ice is measured to calculate the heat

evolved. The extreme difficulty of duplicating exactly the amount, shape, and position of the ice film on the calorimeter at the outset, coupled with the difficulty in causing the film to melt reproducibly, has been a major obstacle in the development of this method. Swietoslawski (*47, 53*) has studied these problems and concluded that the ice calorimeter could be of great value in measuring small, constant heat sources. It has the distinct advantage that a measurement may be carried out for an arbitrary length of time. If secondary effects could be controlled, the accuracy would be almost unlimited.

Shchukarew (*44*) has used phenol fusion in a modification of the ice microcalorimeter.

Adiabatic Microcalorimeters

In adiabatic calorimetry the temperature of the calorimeter jacket is maintained, manually or automatically (*20, 24*), equal to that of the calorimeter itself. In this way an ordinary Beckmann thermometer may be used in conjunction with a microcalorimeter, as pointed out by Swietoslawski and Dorabialska (*48, 49*). In their first adiabatic microcalorimeter, a thermocouple was arranged with one junction at the inner surface of a relatively massive metallic calorimeter. The other junction was placed at the outer surface, where heat was supplied to maintain zero thermoelectromotive force by heating the water bath surrounding the calorimeter. Swietoslawski (*51, 52, 57*) improved and simplified this calorimeter by placing the galvanometer and thermocouple connecting wires inside the jacket and by using a glass calorimeter.

The heat developed by a constant source is given by

$$Q = k(T - T_0) + \Sigma s$$

where k is the heat equivalent of the calorimeter, T the temperature after known time under strictly adiabatic conditions beginning at T_0, and Σs is the sum of all secondary phenomena, including the heating of air inside the calorimeter, the effects of imperfect jacket temperature control, *etc.* If these effects can be maintained proportional to the temperature interval, the accuracy of a comparison method will be limited only by the accuracy of measurement of the temperature difference. The temperature rise can be made as large as is consistent with a reasonable time of measurement. Heat effects of the order of 0.001 cal/hr have been measured.

Zlotowski (*72, 73*) measured the heat evolution of radium in an adiabatic calorimeter of sufficient mass to absorb 97.5% of the gamma radiation. He found a value of 139.6 ± 0.3 cal/gm hr, of which 9.1 cal/gm hr was attributed to gamma absorption from RaB and RaC radiations. This result was confirmed in another adiabatic calorimeter in which the heat source was maintained at the jacket temperature, which was in turn controlled by the temperature of a vacuum-insulated metallic calorimeter placed to absorb a known large fraction of the gamma radiation. The observed value was 9.10 ± 0.15 cal/gm hr.

Zlotowski (*74*) used a very small nickel calorimeter and an improved adiabatic jacket for alpha and beta absorption of RaE and its polonium daughter, finding 1.61 ± 0.03 cal/hr per curie, corrected for polonium activity. This corresponds to a mean beta energy of 0.320 ± 0.005 Mev.

Other literature on adiabatic microcalorimeters includes designs by Lipsett and Maass (*26*), Dorabialska (*10, 11, 12, 13, 14, 15*), Swietoslawski (*50, 55, 56*), Sanielevici (*41*), Conn, Kistiakowski, and Roberts (*6*), and Winand (*69*).

Microcalorimetry from Cooling Curves

If a heavy metallic block is raised to a temperature from 0.1 to 0.2° C above that of a constant temperature bath

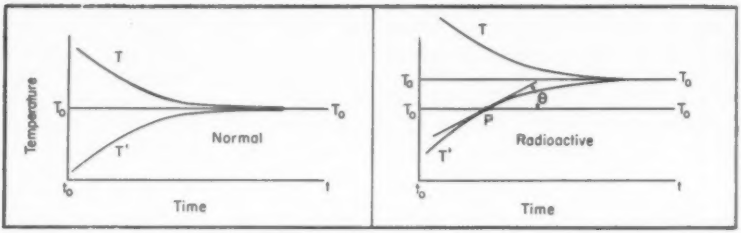

FIG. 5. Cooling curves showing the shift in the symmetry axis for radioactive materials

and then cooled by the bath, the cooling curve (temperature T *vs.* time t) shows no deviation from Newton's cooling law: $dT/dt = A(T - T_0)$, or $(T - T_0) = e^{-A(t-t_0)}$

A heating curve would be symmetrical with the cooling curve, as shown in Fig. 5*a*. If the block were radioactive, emitting heat at a constant rate, the two curves would instead be symmetrical about a temperature T_a higher than T_0, as in Fig. 5*b*. Point P in Fig. 5*b* is of particular interest, since it is the crossing point of the heating curve; the adiabatic condition is applicable. The rate at which the block is heated under strict adiabatic conditions is given by

$$dT/dt = \tan \theta = a$$

The rate of heat evolution may be determined by carrying out heating and cooling curves to equilibrium at T_a, by obtaining the rate of cooling and the rate of heating for a portion of the curve and extrapolating to T_a, or by obtaining the tangent of the angle θ. To eliminate uncertainties in determining heat capacities, the determination may be performed by remeasuring with an additional known heat source.

According to Swietoslawski (*47*) 0.01 cal/hr can be measured with an accuracy of one to two percent. It seems that an experimenter would have to be careful to avoid uneven heat distribution in order to use this method for precise microcalorimetry.

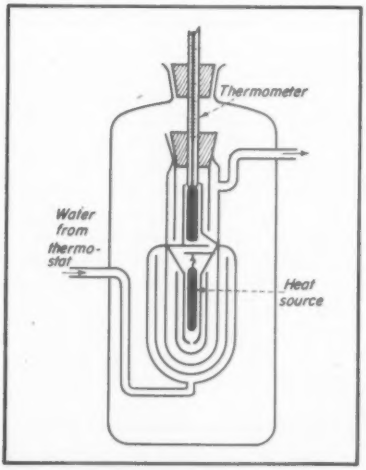

FIG. 6. Swietoslawski labyrinth flow-type isothermal calorimeter

Isothermal Microcalorimeters

Swietoslawski (*54*) has designed a glass labyrinth flow type of isothermal calorimeter in which the temperature rise of the effluent water may be measured by means of a Beckmann thermometer. This isothermal calorimeter is diagrammed in Fig. 6.

The heat emitted by a constant heat source may be found from the relation:

$$Q = vdc(T - T_0)$$

where v is the velocity, d the density, and c the specific heat of the water. Heat transferred from the inner column of water to the second column is not lost, because the second column eventually displaces the first. With several

columns of water the total heat loss becomes negligible. This instrument was calibrated with an electric heater supplying from 1 to 10 cal/hr, and the average error of measurement was 0.5%.

Swietoslawski (*47*) later improved the method by using a metal construction, in which constrictions were provided to smooth the flow velocity. An improved labyrinth further reduced heat losses.

Recent Developments in Calorimetry

Simon (*45*) has performed some exploratory experiments with very low temperature calorimeters. He notes that a lead calorimeter at 1.3° K (obtained with liquid helium) would have a specific heat 0.0003 that at room temperature. A further increase in sensitivity could be obtained with wolfram, which has a higher θ_{Debye} value. At these temperatures the thermal insulation can be very efficient since radiation is greatly reduced, and measurements could be carried out over several minutes. A calorimeter composed of one cc of wolfram might be used to measure 10^{-6} cal/hr with an uncertainty of no more than one percent with available thermometric equipment. For example, the heating caused by gamma radiation of 0.1 mc of radon and its daughters at a distance of three cm could be determined to that accuracy.

By utilizing magnetic methods at temperatures below 1° K it should be possible to extend the sensitivity to measure 10^{-8} cal/hr to within 10^{-10} cal/hr.

Szentpaly (*58*) has reported a small microcalorimeter and thermostat with a sensitivity of 8×10^{-5} degrees C per millimeter throw on an optical scale. The thermostat maintained temperature constant within 10^{-5} degrees C.

Zamenhof (*71*) has designed a surface-tension thermometer capable of measuring temperature differences of the order of 0.0001° C. The shift of the center of the meniscus in a small glass capillary is shown to be a function of only the liquid chosen and the temperature.

An isothermal liquid nitrogen calorimeter has been reported by Stout and Jones (*60*). It was used in the determination of the half-life of Pu^{239} which was cited at the beginning of this article. Plutonium metal was placed in a Dewar flask along with a calibrating heater, as shown in Fig. 7. The flask was attached to a helix of pyrex tubing which led to gas meters. This flask was suspended in a larger Dewar flask filled with liquid nitrogen. The procedure was to condense about 100 cc of liquid nitrogen in the inner flask and then to heat the nitrogen in the outer flask to a temperature slightly above the boiling point, held constant to within 0.1° K. The instrument was calibrated by passing known currents through the calibrating heater and by observing the equilibrium rates of gas evolution. A measurement could be completed in from two to three hours. It was observed that 120 gm of plutonium produced 198.6 cal/hr. The error is probably one percent or less.

Zumwalt, Cannon, Jenks, Peacock, and Gunnis (*75*) used a slightly modified isothermal liquid nitrogen calorimeter to determine the disintegration rate for P^{32}. The validity of a previous value calculated from Geiger-Müller counting measurements was established. A 25-mc source of P^{32} produced 0.0738 ± 0.0009 cal/hr, corrected for Bremsstrahlung.

Recently Jenks, Ghormley, and Sweeton (*23*) have used this instrument in a determination of the mean beta energy of tritium, based on their concurrent determination of the decay constant and on direct weighing of a $T_2 - H_2$ mixture. In one determination 5.17 ml of a mixture containing 2.57 ml T_2 (corrected to 0° C and 760 mm Hg) were adsorbed on palladium,

which was then placed under liquid nitrogen in the calorimeter flask. This amount of tritium was observed to release 0.1909 cal/hr or 1.384×10^{12} kev per second. From the number of tritium atoms, 1.382×10^{20}, and the half-life value of 12.46 years or 3.93×10^{8} seconds, the disintegration rate was obtained:

$$dN/dt = N \ln 2/T_{1/2} = 2.44 \times 10^{11} \text{ sec}^{-1}$$

The mean beta energy was:

$$\bar{E} = \frac{1.384 \times 10^{12}}{2.44 \times 10^{11}} = 5.69 \text{ kev}$$

This value is believed to be accurate to one percent.

A twin-differential microcalorimeter sensitive to 0.001 cal/hr, but relatively insensitive to external temperature variations, has been described by Calvet (*4*). The calorimeter tubes were mounted in a massive copper block. The measuring system included nearly 200 thermocouples, connected in opposition so that a null reading would correspond to equal heating rates in the two tubes.

An interesting departure from conventional instruments is the neutron flux meter of Barbaras, Farr, Kuranz, *et al.* (*2*). It is a multijunction thermopile having alternate junctions coated with boron. The energy released by neutron capture in the boron causes a temperature difference across the thermopile. For slow neutrons the tem-

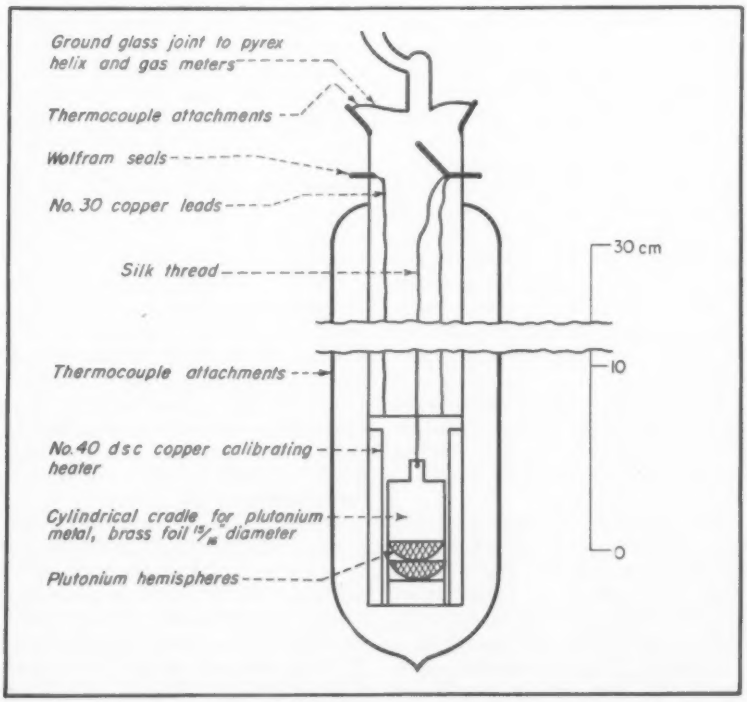

FIG. 7. Liquid-nitrogen isothermal calorimeter developed by Stout and Jones and used to find the half-life of Pu^{239}

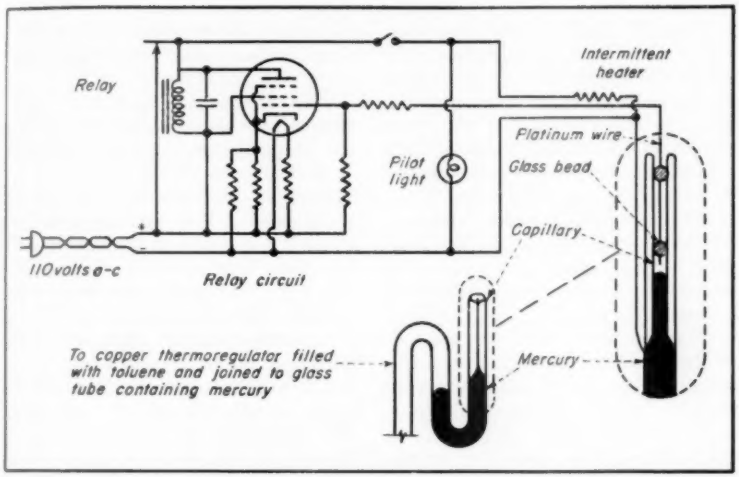

FIG. 8. Thermoregulator relay circuit for temperature control in calorimetry

perature difference, measured by the thermal emf generated, is directly proportional to the number of nuclear reactions.

Sanders (*59*) has described a calorimeter which is particularly useful in the determination of the intensity of a beam of accelerated particles where neutral particle formation is appreciable. In such a case the charge collected is not a significant measure of the energy.

Calorimetry

For a detailed discussion of the design and operation of microcalorimeters, reference should be made to White (*68*), Swietoslawski (*47*), Tian (*61, 62, 63*), and Roth (*40*). Primary considerations are the following:

A constant temperature room is desirable for accurate work with most of the calorimeters which have been used. Temperature variations greater than 0.5° C should not be tolerated. Entrances should be closed with a thick curtain, air lock, or both, and heat should be uniformly distributed.

The calorimeter should be housed in a thermostat, with the temperature controlled to within 0.001° C. This is best carried out by maintaining its temperature slightly above that of the room. Temperature control is most easily and safely provided by means of a large toluene-mercury thermoregulator, made of copper except for a mercury-filled glass neck. As the thermoregulator is warmed, the expanding toluene eventually forces the mercury to contact the platinum point in the capillary. Thus the device acts as a thermal switch and can be used to actuate a relay circuit such as that shown in Fig. 8.

Adequate stirring of the thermostat is important so that temperature variations throughout the water will be negligible. Surface conditions must not be subject to large changes, because these would alter the cooling effect of evaporation. For careful measurements an additional constant heater should be adjusted so that the intermittent heater completes one cycle (of nearly equal *on* and *off* time) in about one minute.

The calorimeter itself should have, in general, a low heat capacity and should

be thermally insulated from the thermostat in order to obtain reasonable temperature differences. These considerations must be balanced against a corresponding increase in the time required to achieve thermal equilibrium.

Conduction losses can be minimized by suspending the calorimeter in the jacket by silk threads and by making a minimum number of electrical connections, using very small gage wires. Air conduction, of course, can be reduced by increasing the gap.

Where practical, conduction and convection losses can be greatly reduced by evacuating the jacket. If a very good vacuum is not employed, measurements will be difficult to duplicate.

Convection losses caused by air currents are particularly troublesome because they generally are not proportional to the first power of the temperature difference. These losses can be diminished by reducing the air gap of the jacket. However, gaps smaller than 1.0 to 1.5 cm are usually subject to appreciable conduction losses. In connection with this, it may be stated that a multiple jacket with about 1.5-cm spacing serves a two-fold purpose. It minimizes convection without making conduction a significant source of leakage, and it smooths out temperature fluctuations of the thermostat to such an extent that, for a triple jacket, variations of 0.001° C outside in a one-minute cycle have no measurable effect on the calorimeter tenperature (*62*, *63*).

Radiation losses can be minimized by coating the calorimeter with a highly polished metal. It should be pointed out here that a noble metal is normally required. The reflectivity of an apparently bright surface may actually be less than 0.85, due to thin oxide layers or other surface conditions. Also, protective coatings such as varnishes defeat the purpose of a reflective surface.

Hoge (*22*) has recently reported that electrical transient effects are held to a minimum if the resistance of the supply circuit is made equal to the resistance of the calorimeter heater.

Calorimeters for alpha or beta measurements should be designed to permit the escape of electromagnetic radiation or to nullify the heating effect of its absorption. A thin-walled calorimeter is advisable. For gamma measurements it is desirable to prevent particle radiations from reaching the massive absorber. Only small corrections need be applied if the source is surrounded by a thin-walled container thermally insulated from the calorimeter block.

The calorimetric method should be employed wherever possible because of its inherent accuracy. The thickness of the sample, degree of backscattering, and resolving time do not constitute uncertainties in its use. Other uncertainties of counting methods have analogies in the calorimetric method, but these uncertainties may be made considerably smaller. As a result no special sample preparation is required.

At the present time, however, the calorimetric method is limited to applications involving relatively large samples. The over-all time spent in making a determination need be no greater than for counting methods, but radioactivities of periods smaller than about one day would introduce serious complications in some of the instruments described in this article, because an appreciable fraction of the activity would decay during the time required to achieve thermal equilibrium.

The possibility exists that a (small) part of the energy absorbed by a calorimeter may not be converted directly to heat energy. The energetic particle may induce phase transitions, chemical reactions, *etc.*, which would result in an apparent lack of energy balance within the calorimeter. Jenks, Ghormley, and Sweeton (*23*) have suggested that such an effect occurs to a small extent in ice at low temperatures.

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The Radiochemical Laboratory—

An Architectural Approach to Its Design

The new field of architecture to which nucleonics has given birth must be approached with revised concepts. How various levels of radioactivity affect planning of labs and offices serves to introduce a proposal for a modular system that offers flexibility in layout. Shielding, waste-disposal facilities, hoods, finishes, heating, and ventilation are touched upon

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NUCLEAR SCIENTISTS AND ENGINEERS have fostered a new field of architecture. Its goal is: The most desirable combination of elements that will provide housing for research, development, and production facilities involving radioactivity. Now reaching adolescence, this specialized field is striving for early maturity with the assistance of architects who must gain the necessary background in nucleonics (*1*).

New concepts must be formulated and assimilated by those planning to go into this work. Take materials used in design, for example. Normally, only four requirements are made of such materials; they must:

(*1*) Resist such forces as may be brought to bear on them
(*2*) Withstand climatic attack
(*3*) Be readily fabricated
(*4*) Where necessary, appear pleasing to the eye.

In "nucleonics" architecture, however, there are three additional requirements; the materials must also:

(*5*) Resist severe chemical attack
(*6*) Provide shielding from radioactivity
(*7*) Be decontaminable and/or replaceable with facility.

Four factors have grown to seven, and we have considered only the properties of materials. With this, then, as a rough proportion, it is easy to see how complications multiply when one considers the various aspects of radioactive laboratory design.

General Planning

After determining what elements must go into the final plan and layout, the architect specializing in this field must think next about grouping the elements with respect to the level of radioactivity associated with the area.

Three main factors guide him in determining the permissible levels in a given area: effects on radiation backgrounds in juxtaposed areas, health hazards, and extent to which safeguards must be provided to control the two foregoing. Levels of activity, functions, and facilities included in each, can be summarized as follows (*2*, *3*):

1. **Sub-normal:** Precision work of special types, and counting rooms.
2. **"Cold," or normal,† activity:** Offices, libraries, conference rooms,

* Member, Amer. Inst. of Architects.
† No greater than background due to natural radioactivity.

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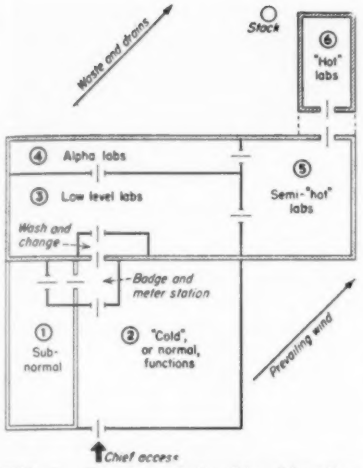

FIG. 1. General relationship of location for the various levels of radioactivity

eating facilities, "regular" shops, storeroom, *etc.*

3. **"Low Level":** "Warm" shops and storerooms, laboratories for β and $\gamma \sim 10$ mc, and $\alpha \sim \mu$c.
4. **Alpha:** Well ventilated laboratories for $\alpha > \mu$c.
5. **"Semi-Hot":** Well shielded laboratories for β and $\gamma > 10$ mc, but < 1 c.
6. **"Hot":** Areas housing cell blocks and caves for β and $\gamma > 1$ c.

Some special operations of the highest radioactive levels must be placed well to leeward, and/or be separated by a low ridge.

Sub-normal level work must likewise be removed, but in the opposite direction, preferably to windward of "hot" work. Provision must be made to prevent any radiation being introduced into, or contamination of, such areas. Normally about 2 ft of concrete will be needed to shield such spaces, with access gained through a labyrinth to keep out all extraneous radiation that might raise the background above the permissible limits for instruments within. These areas should be as near zero activity as possible.

The level of cold, or normal, activity is where most of the administrative and service elements will be situated. Activity here may be slightly above normal background. Eating facilities, however, must be kept scrupulously clean, especially with regard to α radiation which is not very penetrative and therefore more difficult to detect than β or γ radiation. It is through the cold area that all access must be controlled to other levels (*4*). Frisking devices are a requisite for this area to detect any radioactive contamination on persons coming from areas of higher activity. The ideal juxtaposing of all the elements is indicated in Fig. 1.

In the low-level area, where the bulk of the tracer work is located, background should be kept so low that the sensitive counting devices in rooms adjacent to the labs will not be affected. Juxtaposition of semi-hot labs with low-level work may be necessary; in such a case the problem of preventing the introduction of activity is magnified. Doorway detectors and probes may be necessary to prevent spread of contamination from the semi-hot lab in this area to adjacent quarters. Only the cooperation of personnel can effect contamination control. Facilities must make good procedure easy, and faulty ones difficult.

The alpha area requires air locks to eliminate possible spread of α contamination to adjacent areas. No shielding problem is presented, however, because of the short range of α particles. Great care must be taken to keep all office space in this section free from contamination.

The semi-hot facilities will all need about 2 ft of concrete around labs and decontamination rooms, in addition to the local shielding which will be provided in the form of lead bricks to protect operators. Here, again, there will

be offices and small counting facilities adjacent to semi-hot rooms; care must be taken that contamination does not raise the background level of these quarters.

Hot facilities will require enclosure within some 3 ft of concrete, in cells or caves (*5*). See Fig. 2. All operation will be by remote control, and carefully designed vents and drains will keep contamination from escaping the blocks and entering the work areas surrounding. This section will be as far removed as feasible from the sub-normal area.

It will be seen from Fig. 1 that one should go from one level of radioactivity to another in progressive (or regressive) increments. A health physics checking station, centrally located, would control all ingress and egress. Portable, personnel meters would be picked up here for required use in the more active areas, and permanent frisking devices would check personnel in such manner as seems advisable. Adjacent would be wash rooms and clothing change facilities for personnel decontamination.

Naturally, one cannot hope to adhere strictly to this layout of activity levels; it is schematic in nature. For example, in the hot area, some offices and record rooms may be needed. Or it may be necessary that some semi-hot labs lie directly adjacent to the hot labs. It is up to the architect to satisfy the needs of the function to be housed, the health physics demands, and the requirements of over-all design.

Opinion is at present divided on whether all levels of activity should be incorporated into one building, or whether each type of activity should be located in separate buildings connected with a covered passage. Both views have points in their favor. The one-building scheme would reduce construction costs, minimize travel distances, and permit flexibility where space is concerned. However, in case of an accident or explosion in a hot lab, it is possible that much of the building could become contaminated before emergency doors and other safety features could function. Perhaps a great part of the structure would become contaminated (usually by foot traffic) beyond hope of satisfactory decontamination operations. Much time would be lost in demolishing and rebuilding. In separate buildings, the initial cost would be higher, but if an explosion or a long-term gradual building up of background made one of the structures too hot for safe occupancy, it could be cleaned or abandoned without affecting adjacent areas in an undue manner. For the present, however, let us hold to the premise that all elements will be placed in a single structure.

The areas indicated in Fig. 1 may apply to grouping of separate buildings as well as a single building, but as far as possible the general interrelationship of facilities and direction of prevailing wind and hot waste disposal should be maintained as indicated. The wind will then carry any escaping vapors away from cooler areas toward warmer ones. Stack gases from the hot areas will continue to dissipate also in the direction away from the cooler areas. Research currently in progress should assist in future elimination of practically all activity from these stack gases. Liquid wastes, where a hot drain system is provided, will likewise flow in this direction so that vapors from evaporators and settling ponds will not tend to drift back toward the operation and administration areas (*11*).

Vertical Aspects of Design

Sub-normal level. Two-foot thick concrete walls and roof slab will be needed to protect sensitive recording and counting instruments from radiations that would tend to raise the background level in this area. If housed in

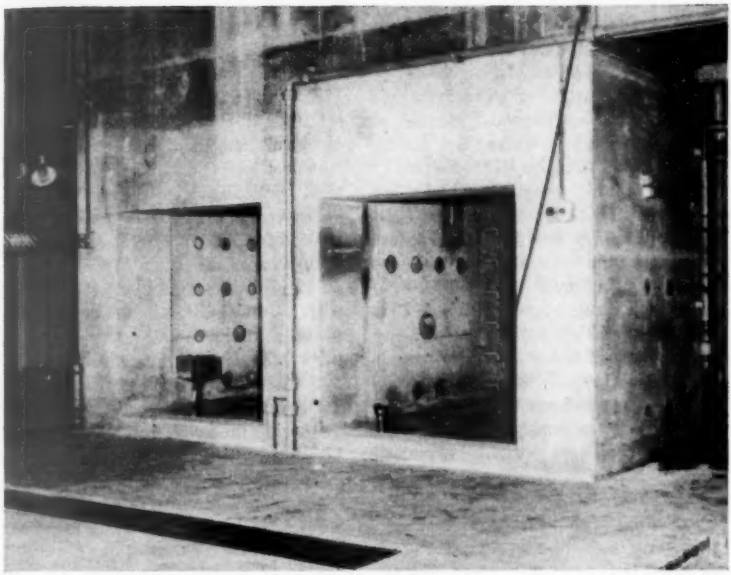

FIG. 2. Concrete cells on the first floor of an Oak Ridge semi-works facility. Both inner and outer thick concrete walls have 4 and 6-in. access holes with removable concrete plugs. Floors around cells are pitched to drainage trenches, lines of which go to the process-waste tank*

a single structure and placed below the ground; this area could then serve as a foundation for office space above, or for labs dealing with low activities. In any event, the sub-normal area must be well air conditioned; temperature and humidity may need to be controlled with utmost precision ($\sim \pm 5°$ F and $\pm 1\%$). All incoming air should be filtered with efficient filters to remove the major portion of dust particles.

Normal level. Little deviation from standard construction practices need be observed. True, prevailing wind and drain directions must be kept in mind, but vertical placement of facilities is possible in this area, and one need not be limited by the number of stories.

Low level and alpha activity. The facilities in these areas may be placed on several floor levels, if necessary. The chief problems to provide for are proper shielding of hot drains and exhaust duct from β and γ sources in area 3, especially when the drains and ducts originate above the ground floor, and careful evacuation of α contaminated vapors from area 4 labs. Counting rooms can, for convenience, be located in these areas, but may need 2-ft concrete walls and ceiling (also, floor, if facilities below it involve radioactivity). In general, it is desirable to locate area 3 facilities and counting rooms at grade level, for at that level there will be motorized dollies with heavy lead shielding and much local shielding employing lead bricks. Therefore, any counting rooms placed over these labs might need a 2-ft floor slab. Shielding of hot-drain vertical runs is also simplified when their point of origin is not

* Photograph from "Problems in Planning Facilities for Radiochemical Process Development" by A. C. Jealous, AECD-2512 (1949)

above grade; likewise, exhaust from hoods, if led downward, would then need less protection. The tendency now seems toward overhead service and air supply, with downward exhaust of contaminated air as well as hot drains.

The α labs may be located upstairs with less trouble, especially when using dry boxes for this work, rather than hoods. The dry box (discussed later) requires less air for ventilation than the hood by a factor of about 100. No shielding is needed for α drains and air exhaust; the chief problem is to insure against leaks of either liquid or vapor.

Semi-hot level. Here all labs, the entrances to which may be through labyrinthal passages, must be shielded with 2-ft-thick concrete walls. If poured concrete is used, a multi-story building may be constructed with little difficulty, but flexibility would be sacrificed. In such a structure, however, if only semi-hot work is to be done therein, the entire floor area would have to be designed to take heavy equipment, or thick walls, at any point throughout. More preferable is to make use of a ground floor and construct 2-ft-thick interior walls of solid concrete block (some sites are considering aluminum block) that may be moved without undue cost when variation in layout is necessary. This flexibility is most desirable.

Hot level. A separate one-story building is the only type of structure appropriate to house the cell blocks and caves (whose walls must be of concrete about 3 ft thick) required for "hot" work. Certain service functions, clothing change, equipment decontamination, offices, and attendant labs could be grouped about the main work area in one or more stories, depending on the function and its use.

Flexibility and Module

The factor of flexibility has been mentioned as most desirable. This can be attained by adhering closely to a modular system of planning. To expedite consideration of modules, let us accept the premise of a $1\frac{3}{5}$ factor (see Fig. 3). The structural system will be set up on 12-ft (or 24-ft) column centers, along the length of the building, with windows likewise located on 12-ft centers; vertical service risers, too, would be spaced on this 12-ft module. The subdivision of 3 ft would place lighting fixtures accordingly, and the windows on 12-ft centers would be 5 ft-6 in. wide with a 3-in. mullion in the center. Interior partitions could then be 3 in. thick, movable, steel panels which could be located anywhere along the length of the building on 3-ft centers. Whether we decide on a $1\frac{3}{5}$ module, or one of $1\frac{3}{4}$, $1\frac{5}{3}$, or $1\frac{5}{5}$ proportions, is of little interest here. We shall deal further with this when considering lab equipment. The important thing to remember is that uniform locations for service risers and movable interoffice and interlab partitions are necessary to achieve flexibility.

In such a new field as radiochemistry, it is hard to foresee precisely what calls the future will make on equipment and layout. Requirements will always be changing; one must be ever ready to alter sizes, shapes and positions of various functions to handle new problems with utmost facility.

The width of the structures will vary. However, the interior module of 3 ft should aid in determining width precisely. If panels of 3 ft (4 ft or 5 ft) width are to be used, the final clear dimension between hallway (or service riser) and exterior walls should be in multiples of this figure. So long as this factor is maintained, and provided a standard clear ceiling height (of, say, 10 ft) is used as a premise, widths are of little matter (except structurally), and all movable partitions will fit neatly into any building or wing thereof. Naturally there will be exceptional cases where special treatment will be de-

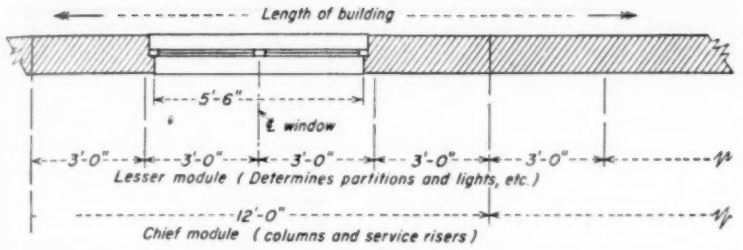

FIG. 3. A modular planning premise

manded. See Fig. 4 for typical lab and office layouts.

In the case of semi-hot labs which call for shield-partitions of 2-ft concrete thickness, the solid block of 4 × 8 × 16 in. with little or no mortar can be used as a movable partition. It must be accurately square, of uniform dimension, and have a precisely smooth surface. These qualities further reduce possibilities of penetration.

It is sometimes argued that a preferable type of block is one with ridges that interlock with grooves to cut down on any collimated radiation that may come through the horizontal joints (vertical joints are staggered). It has been found at the Oak Ridge National Laboratory, however, that such an occurrence is extremely improbable. In general, with a source directly in line with the crack, the "beam" is so scattered by the time it comes through the 2 ft thickness that no dangerous level of radiation is present.

Contamination Considerations

Shielding. General shielding of area 5 is covered by the foregoing, but special shielding within hoods is considered here for areas 3 and 5. As many as 3 tons of lead bricks are sometimes used to build up the shield in a single hood. Usually, four hoods will be in a single lab, so that one must keep in mind for floor loads in these areas roughly 10–12 tons of lead brick in use and/or storage.

In areas 4 where γ radiation and massive shielding are not the factors, the problem is of another nature: α contamination. Here efficient removal of contaminated vapors is needed. Thin, *tight* shielding is required to control active exhaust gases until they are cleaned and exhausted.

In areas 5 and 6, or for other special needs, unique shielding methods may be called for. Charts are available that indicate "what" thickness of "which" material will be needed for "how" active a source of various radiations to reduce the activity to a specified level for personnel or instruments (*6*). The Health Physics Division of the AEC can provide this data.

No further shielding is needed for area 1 activities beyond the encompassing 2 ft of concrete, except for such protection as may need be incorporated into regular design of instruments and containers used in this area.

Finishes. On the matter of materials for construction, let us look more carefully at the subject of partitions. It has been pointed out that solid concrete blocks are more desirable than poured walls for reasons of flexibility. Let us think now of decontamination in a hot lab. Whether one uses block or poured concrete, there must be a finish of some kind, painted or sprayed on the surface of the walls.

One finish recently suggested calls for about 2 coats of Prufcoat, Amercoat, or

FIG. 4. Typical laboratory and office layout

equal, followed by a special coating of stripper base, over which a strippable surface may be painted or sprayed. When such a surface becomes contaminated, the strippable coat may be peeled off. Should activity have gone through this, the base may be removed with a dilute alkali which would not disturb the underlying paint. Even the latter may be removed, if necessary, by softening with a solvent prior to scraping off. Finally, where surfaces have been damaged prior to a major spill or explosion and activity is permitted to contaminate the concrete itself, it can be seen that replacement of blocks will be much more facile than chipping or breaking out a poured wall.

Likewise, for movable metal partitions for areas 3 and 4, permanence is a far less important asset than decontaminability. In cleaning or moving a tile or plaster wall, much dust is generated which is highly undesirable in the vicinity of radiochemical work. Metal partitions may be sealed at the joints with narrow strips of polyethylene and covered with the peelable paint combination mentioned above for concrete. Usually, just the first two layers of paint will suffice; the stripper can be omitted.

Peelable surfaces are needed only where contamination from spill, spray, or explosion is possible. Elsewhere, Prufcoat, Amercoat, or equal, has been found satisfactory for decontaminability when such surfaces may be less severely exposed to possible hot "baths." The exterior surfaces of cells blocks, caves and walls in rooms where high activity is closely confined, and well controlled, would be suitable for treating in this manner.

Actually, it was found recently that, although representatives from most laboratories favored the use of peelable surfaces, few had then employed them and, of those who had, none had exercised the peeling properties because no serious spills had occurred. However, all agreed that theoretically this was the most promising of possible surface treatments for walls. Further application of this treatment has since been found satisfactory at Oak Ridge.

For floor covering in areas 3 and 4, asphalt tile has an advantage over most other coverings. When laid on felt, or some more impervious material, with a mastic binder, there is little danger of contaminating the concrete floor. The tiles must be firm enough not to compress excessively under dolly weight, yet not so hard they will not tend to "flow" when in use and help seal cracks between tile. A serious spill would probably result in some contamination of the undermat. By cutting out the contaminated section of tile, removing part of the undermat, and overlapping a clean piece, however, fresh tile may be laid into place. Linoleum presents an uncracked surface initially but is more difficult to match neatly after cutting out a contaminated section. Most other floor coverings tried to date have been found too expensive or too porous and difficult to decontaminate, and/or replace.

In areas 5 and 6, there is usually such heavy equipment to be moved about that the most desirable floor finish is the concrete slab itself. A good surface hardener should be used to avoid porosity as far as possible. Finishing with Prufcoat, Amercoat, or equal, seems the most acceptable method of painting; it stands up well under traffic. Should a spill occur which the usual scrubbing will not decontaminate sufficiently, the concrete may be broken out 2 or 3 inches deep, and a new finish poured. It is argued in some quarters that the heavier the traffic, the less need there is to paint concrete floor surfaces at all.

Structure. The structural system itself, for radiochemical design, offers no unusual problems except for the heavy loads for which provision must be made;

as has been suggested, in most cases all heavy elements can be held to the ground floor. There will, of course, be no bearing walls except possibly the exterior shell, as all space inside will be "universal" and broken up for use by adjusting the movable partitions.

A point one must keep in mind when designing radiochemical facilities is to avoid any pockets or ledges where dust may gather. Such places could build up so much activity that readings on instruments would be affected or permissible level for humans surpassed. Cracks and crevices are equally dangerous, and even harder to decontaminate without losing much time.

Service Supply

Services coming into the labs should feed from overhead. Access to them will then be better for two reasons: (1) The space below the grade on which the labs rest will have hot drains and ducts; these exhaust ducts and drains will be under corridors. (2) Above the lab will be sufficient plenum to introduce air supply and provide space for incoming vapor, liquid, and power supply. See Fig. 5.

As previously mentioned the service risers will be located on a modular basis, every 12 ft. Referring back to Fig. 4, we see there service riser spaces ($3\frac{1}{2}$ ft $\times$ 7 ft) enclosing the columns. Each service space would not, at all times, be in use; valved T's would be provided in the plenum to make connections as they are called for. Using the movable metal panels (or solid concrete block with access doors) to enclose these risers, access may be had from all four sides. This is the chief advantage to be gained over placing risers on the exterior wall, where usually but one side is accessible for making connections or repairs. Between each service riser is a space about $4\frac{1}{2}$ ft wide which can serve either as entrance to office or lab, or as closet or alcove space for the corridor or the adjacent room. In this manner, services are readily accessible, no space is wasted, complete flexibility is attained, and corridor widths may be held to a minimum because no doors will project when open.

Horizontal service runs, from risers to equipment, should be kept accessible, yet be covered to prevent dust accumulation. The best location is on the surface of metal or concrete block partitions, under the counter tops of lab equipment. Piped services would be available only along interior partitions

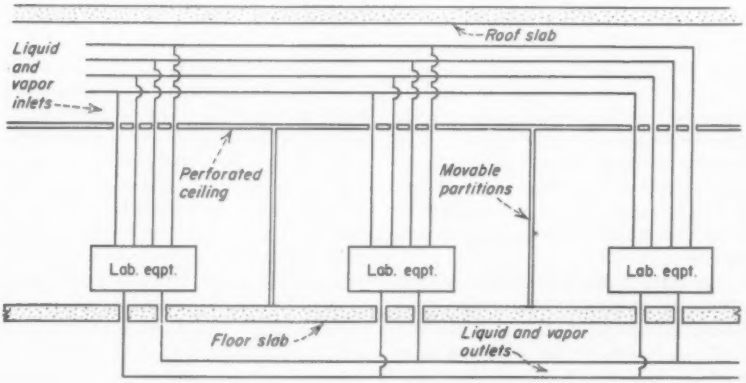

FIG. 5. Schematic diagram showing services

or from peninsulas running from them. No piping should be run inside walls or under floors because access would involve loss of time and excessive dust. Electric service outlets may, of course, be placed along the exterior wall, as well as wherever else needed.

Flush-mounted, fluorescent fixtures are the most desirable as a light source in the ceiling.

Heating and Ventilating

Air for ventilation in labs must have controlled temperature and humidity, both winter and summer. Based on an average of four hoods per lab, each calling for about 1,000 cu ft per min, the total demand for air for each lab will be about 4,000 cu ft per min. In the winter, air temperatures may be kept down by the use of radiant heating panels in the floor or ceiling and within exterior walls. For air inlet, to take care of such great air demand, aspirating ceiling diffusers are not desirable; perforated sections of the ceiling provide more satisfactory supply. Solid sections of equal dimension (say 1 ft square) may be used to interchange with the perforated units to balance the air inlet from above. Aspirators induce far too drafty a source of air for use in radioactive labs.

Air used in offices *may* be recirculated, but that from labs *must* be cleaned and disposed of, as will be described later in further detail. All air should be filtered with an efficient filter prior to use to remove any active particles brought in by shifts of the prevailing wind. Likewise, much time and material can be saved by elimination of just plain dust and dirt; it wreaks havoc on operations and equipment in laboratories.

After being conditioned, fans will push the air through ducts to points of use. Theoretically, the ideal circulating system would first take the air to offices, from which it would pass

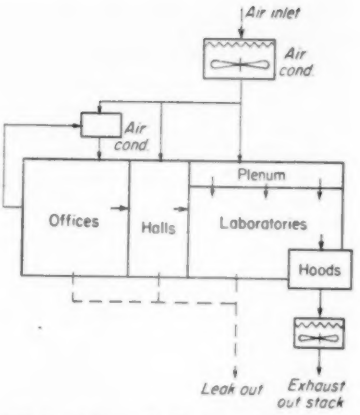

FIG. 6. Schematic diagram of ventilation

through corridors, on into the labs. It would then evacuate via the hoods. Such an ideal scheme is indicated roughly in Fig. 6.

Offices should be operated slightly above static pressure, corridors at about the same level as outside, and labs slightly below atmospheric pressure.* There will be then little tendency for contaminated vapors to drift back through the rest of the building. One new factor that will assist in maintaining this correct balance of pressure is discussed under the subject of equipment.

In regard to the placement of radiant heating panels in either the floor or ceiling, P. B. Gordon said recently: "Since most of the rooms will be built on grade with concrete floor slabs, I believe the *most likely* interior building surface to be used as the heating radiator would be the floor. As a rule, the cost of installing floor systems is less than that of installing ceiling systems, when all the costs are included." He went on to say, however, that a ceiling panels permit operation at temperatures "as high as 105° F to 115° F as com-

* These differentials are approximately fractions of an inch of water.

pared with a top limit of 85° F for floor panel operation."

Current thinking on the subject, therefore, leads to the following summary: Ceiling panels are the most effective, along with exterior wall radiant heating panels, as less linear footage of pipe is needed. Imbedded floor piping is the easiest to install, but the time lag is much greater in controlling temperature, and more linear footage is needed because of the lower temperature at which the water must circulate. Individual factors at a specific site will determine which type, or combination thereof, to adopt.

Equipment

Equipment of a permanent nature for use in labs is restricted chiefly to cell blocks and caves. Poured concrete walls about 3 ft thick will provide adequate shielding. The exterior surface should be painted as mentioned to facilitate cleaning. The interior should be surfaced in such manner that decontamination will be possible for whatever type of operation is carried on inside. In some cases a completely welded lining of stainless steel No. 347 will be needed; in others, a bonded plastic lining, or even coatings applied by brush or spray, may suffice. In any event, preliminary decontamination must be carried out by remote control, followed by more careful cleaning manually before personnel can work for any extended period of time within the cell or cave. Numerous peep holes (see Fig. 2), periscopes, telescopes and remote control devices will need to be incorporated into the design of this permanent equipment for hot work (*2, 3, 4, 5*).

Semi-permanent equipment will be in use in area 5 for semi-hot work. Tables, benches, hoods, and vaults make up the bulk of such equipment in this area. The term "semi-permanent" is used to indicate that such an item is normally located in one place, but that it may be disassembled in order to be moved, serviced, decontamined, etc. Lab tables and benches are no problem; they should, however, be of metal with shelf and cabinet units below the work surface.

The services should be brought up under a raised ledge at the back of work surfaces, so that they may be carried horizontally through the riser surface at the front of this raised step. If brought vertically through a horizontal plane, active materials would tend to run down quite readily through such a connection and contaminate below the work surface. Spigot outlets should not point directly toward an operator; rather should they point diagonally downward in a plane parallel with the wall, or piece of equipment. This will prevent splashing or spraying personnel should a hose slip off the spigot.

The material most desirable for the work surface is stainless steel, but a peelable finish to cover other material, as described in connection with movable partitions, would probably suffice where abrasive action is not predominant. Stainless steel trays are useful and simplify decontamination.

Hoods in semi-hot labs are not quite so readily made "semi"-permanent as are benches and tables. However, sections can be made demountable to assist in decontamination procedures. The base should be massive to protect γ radiation from going downward toward the operator and to provide sufficient support for some three tons of lead bricks which may be used for shielding activity within the hood. The work surface might slope back toward a cup or ditch in the rear of the hood to prevent spread of contamination when cleaning this surface. Sliding doors must be provided to close the hood when no operator is manipulating therein, and all services should be operated from outside the hood door.

Mirrors will be needed to see what takes place behind the lead barriers. Stainless steel No. 309 or 347 is best for the work surface as well as the vent baffle inside the hood and all of the superstructure. For ease in decontamination, a removable stainless steel work surface and baffle are desirable. Hood lights must be so located that glare is minimized and access is available from outside the hood to service the electrical equipment. A concrete or lead storage cave will be needed in each lab in which to keep active samples ready for use in the hoods.

Hoods for area 3 can be more movable than those described for area 5. They may be built in two sections. See Fig. 7. The upper part includes the hood working surface. The complete assembly can be removed for servicing and complete decontamination by merely disconnecting services prior to dislocation with a hoist. The lower part consists of a secondary lab table work surface; below, if needed, a readily replaceable exhaust vapor filter may be located. The filter placed at the immediate point of vapor exit from the hood reduces the rapidity of contamination build-up within the exhaust duct system and will save expense in later maintenance.

The complete assembly has a constant demand (whether or not hood doors are open) of about 1000 cu ft air/min. A constant velocity of air through hood doors in any position would be about 130 lin ft/min. These conditions can be maintained in the following manner. Hood doors slide horizontally in opening—engaging gears that activate by-pass shutters; this permits the 1000 cu ft of air demand to be satisfied by a vent by-pass when hood doors close, and vice versa. At all times, the call for air intake will be so proportioned that demand through the hood doors will remain constant at 130 lin ft/min. In this way the operator has sufficient ventilation for his work without excessive drafts created by slightly opened hood doors, and the problem of air pressure balance between labs and corridors does not arise because there is always a constant demand for air as initially provided in design (*7*, *9*).* (With conventionally vented hoods, as used in the past, two adjacent laboratories, each with, say, four hoods, could build up considerable pressure differential, should all hood doors be closed in one room, and all open in the other.) In addition, when a by-pass is located near the floor, its action will serve to reduce the activity background level in the room; hence, the adoption of a down, rather than up, draft premise. Services come in at each end of the hood, fed through the vertical end panel; spigot outlets point backward and downward at a 45° angle.

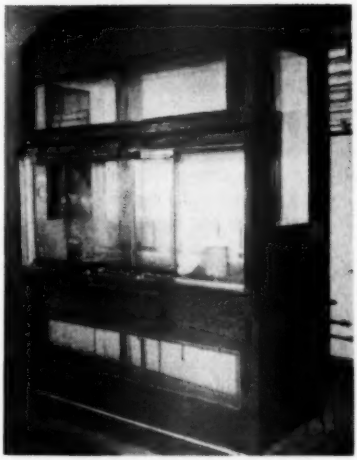

FIG. 7. This mock-up, used to develop hood described in text, shows gears (exposed for picture), rack on under edge of doors, weighted chains for lifting by-pass shutters, secondary transparent access panel above doors, and diagonal box for housing source of illumination

* NOTE ADDED IN PROOF: New developments indicate an additional saving of 50% of this indicated air demand by hoods can be effected.

Dry boxes, for α work, are more desirable than hoods, from the engineer's viewpoint, in that the ventilation needed is less by a factor of 100; only 10 cu ft air/min need pass out through the exhaust vent. Here again, a replaceable filter should be provided at the exhaust connection with the dry box, to eliminate the major part of contamination in the duct system; no bypass is needed. Dry boxes can be very light weight, but *must* have *no* leaks in the exhaust system. (Operators, for the most part, realize that all α operations should be carried on in a dry box, but some use a hood because it is "more convenient" despite the fact that less safety is provided against contamination and more cost is involved because of the greater demand for air by hoods.)

Equipment in general will need to be thought of in modular terms. To derive the utmost benefit from the universal space when subdivided into lab and office areas, the dimensions of equipment must be correlated with the spaces into which it will be installed. With this thought in mind, it is of little import "which came first": the building module or the equipment module; they should, however, be interrelated. Likewise of prime import, is to standardize the design of equipment so as to maximize interchangeability of units of similar function when repairs are needed or lab layout is to be revised. Many standard items now on the market will serve admirably, but a large proportion of the equipment needed in radiochemical laboratories will have to be developed from scratch. The architect will continue to function here in his normal capacity as coordinator, interpreter, and mediator between the client, the engineer and the constructor to effect the most desirable equipment design.

Waste

There will be at least three types of liquid waste, of which disposal must be made: Sanitary, Process, and "Hot." The sanitary system will not differ from that required for the normal disposal of waste liquids. There will be outlets of this type in all areas for non-chemical, inactive needs.

The process sewer will be for the usual chemical waste. Nothing active shall be disposed of via this route (microcurie quantities would not, however, be of any danger, if drained into this but occasionally).* Liquids in this system need only to be neutralized, prior to disposal as ordinary waste.

Hot drains (*5*) will come from cell blocks, hot hoods and sinks, storage lockers and caves, and any place where it is likely that contamination will be at the millicurie level, or higher. These drains should be shielded within the building, and terminate in underground stainless-steel tanks just outside. Here, the acid wastes will be neutralized before being checked for type of activity, and sent to large tanks or vats for permanent storage or disposal (*10, 11*). Maintenance of this drain system is a potential problem and any leak is bound to result in highly radioactive contamination. Therefore, it should either be completely maintenance-free or accessible under such conditions that the probable resulting contamination will not be serious. At no point where corrosion of the line is likely to be a problem should these hot drains be buried in walls or slabs. It is for reason of this needed free access to such drains that all services should be fed from above, as previously indicated. Clean supply lines should not be crowded into an area where hot exhaust drains and ducts are located.

All labs located in areas 3, 4, 5 and 6 need connections with both process and "hot" drains. Normally, process lines will come from sinks outside of hoods or cell blocks, and hot drains will be from

* See "Interim Recommendations: Disposal of Radioactive Wastes by Non-AEC Users," NUCLEONICS **5**, No. 2, 22 (1949).

within hoods, cell blocks, caves, decontamination rooms, *etc.* In either event, stainless-steel, or lead-lined, sinks will dump through a Duriron trap into the respective systems. In less pretentious facilities, which have no hot drain, active material may be collected in a sink (with process drain connection), and aspirated out into a shipping container in which to be kept until transportation to a point of disposal is effected. Activity of the solution used for cleaning the sink should be sufficiently low to further dilute and flush down the process line, provided the batches are $< \mu c$ level of activity, and not dumped frequently.

Exhaust ducts emanating from labs and hoods where β and γ activity are present may need some shielding to protect personnel, especially maintenance workers, who will be in close proximity when making repairs, decontaminating, or altering the installations. The problem of shielding will be nowhere near as acute if filters are located in the outlet at each hood, as previously indicated. Ductwork must be finished on the inside with no joints or rough spots wherein active material might gather and build up "hot" pockets. The "hot"-duct problem has two solutions: (1) Weld heavy stainless steel sections together to provide a "permanent" solution. (2) Use light, inexpensive materials which may be replaced readily. The answer to these two quite opposite "solutions" may be a median approach.

Ducts may be led to a common point of further filteration, washing or electrostatic precipitation of dust particles, after which, the vapors should be forced briskly up a high stack for exit and dispersion. For economic, as well as aesthetic reasons, it is desirable to manifold all exhaust ducts possible in order to have but one stack or roof monitor; the minimum number of stacks or vents should be an objective, keeping in mind prevailing-wind direction when locating, to avoid reintroduction of active particles into any work areas. Current studies are being directed not only toward an almost perfect cleansing of waste gases prior to their being eliminated through the stack but also to various aspects of meteorological significance.

Conclusion

Each of the levels of activity requires a different set of postulates with which to deal when considering design needs. But, by striving for *universal space* and *standard items of equipment*, more desirable facilities can be provided at less cost.

Flexibility, too, is attained by, for instance, movable partitions and equipment which offer personnel safety yet facilitate space alterations and provide easy maintenance and decontamination. Although initial costs may be no lower than a "permanent" type of installation, future maintenance costs are considerably reduced.

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Microchemical Methods Used in Nuclear Research

Microchemical methods developed primarily for use in nuclear chemical research are described and discussed in detail.

Techniques described include microweighings, micro volumetric and gravimetric analysis, solubility measurements, methods for the study of reactions between solids and gases, the preparation of small samples of various reactive metals, density measurements, microcalorimetry, microspectrophotometry, magnetic susceptibility measurements and vapor pressure measurements.

In addition, semimicromethods developed for use in the investigation of nuclear reactions are discussed.

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MICRO AND SEMIMICROMETHODS have found extensive application in AEC and other laboratories engaged in nuclear and radiochemical research. The principal applications have been in connection with isolation and investigation of properties of synthetic and other rare elements. Of comparable importance, however, has been the widespread use of semimicromethods for chemical separations required in investigations of various nuclear transformations. In the latter cases, small-scale methods are preferred because of their superiority in speed and efficiency as compared with the common macro procedures.

An additional factor recommending the use of micro or semimicromethods for research with radioactive materials is that adequate protection for research personnel is more readily effected by confining hazardous operations to relatively small areas.

No general review of micro methods and apparatus developed for use in nuclear chemical research has appeared since Seaborg's (*1*) brief description was published in 1945. It is the purpose of the present paper to review these developments in somewhat greater detail and to bring the subject up to date as far as is possible.

The primary justification for micromethods is to be found in their application to the investigation of rare materials. Prior to 1942 these applications were mainly in the fields of organic and biochemistry. With the production of the first few micrograms of plutonium by cyclotron bombardment in June of 1942, it became necessary to develop a broad field of inorganic micro methods suited to submilligram quantities. This development was initiated by the microgram scale isolation and determination of the specific activity and half-life of the isotope Pu^{239} (*2*). There followed a rapid development of methods suited to the general investigation of the properties of plutonium and its compounds.

At the end of the war, when plutonium had become available in substantial amounts, there was keen interest in investigating the chemical properties of other synthetic and rare elements, which in some cases were of more limited availability than had been

the case with plutonium. Existing micro methods were refined and new methods devised for the purpose of investigating these elements.

In many cases, techniques and methods worked out before the war were of great value in providing a foundation for the specialized developments required by the various wartime "Projects" and AEC laboratories. Of most immediate application to the problems involved was the work of Benedetti-Pichler and Cefola (*3*, *4*) and their co-workers and of Kirk and his co-workers (*5*).

The principal developments in the field of microchemical apparatus and techniques designed primarily for research in nuclear chemistry have been the following:

MICROMETHODS (Microgram to Milligram Scale)

Microbalances

Commercially available microbalances have been used but little in investigating the chemistry of the synthetic elements, since these balances are, in most cases, insufficiently sensitive. Three types of elastic microbalances have been developed especially for work of this kind. The general design of these instruments is shown in Figs. 1–3. In no case is the design wholly novel, since instruments of somewhat similar construction and comparable sensitivity have been described previously (*6*, *7*, *8*, *9*).

At comparable sensibility (defined later), the three types of balances illustrated differ widely in load capacity. At 0.01 μg sensibility, for example, the load capacity of the cantilever balance is about 1 mg, of the simple torsion balance about 25 mg and of the torsion balance with vertical suspension about 5 gm.

The cantilever or Salvioni balance is by far the simplest and cheapest to construct. A balance of this type was used by Cunningham and Werner (*2*) for the first weighings of pure compounds of plutonium. The design shown in Fig. 1 has since been employed by the author and Koch (*10*) for weighing small samples on the milligram, as well as microgram, scale. An instrument of this type may be constructed for less than $100.

The cost of construction of the torsion microgram balance of the Kirk-Craig-Gullberg (*11*) design varies from about $2,000–$6,000,* depending upon the degree of refinement of the various mechanisms used for the beam release, pan arrestment, optical system adjustment, *etc.* A number of these instruments have been constructed for use in various AEC laboratories. Their general operating reliability is excellent and they are distinctly superior in speed of weighing as compared with the conventional knife-edge microbalance. They are also less sensitive to environmental disturbances, such as vibration or changes in room temperature.

The Garner (*12*) balance represents a more recent development in the direction of providing an "ultra micro" balance with a load capacity comparable to that of existing knife-edge balances. One model of this instrument tested by the author showed a more pronounced tendency to drift than did a Kirk-Craig-Gullberg balance under the same environmental conditions. The Garner balance would appear capable of improvement in the design of the pan wells, which are insufficiently insulated and a source of thermal disturbances after being handled.

* A modification of the Kirk-Craig-Gullberg balance, designed by the author, has been constructed for about $500. The modified balance compares favorably with the original model in reliability and ease of operation.

Because of the large inertia of the loaded balance system, the Garner instrument must be manually damped after loading to stop the prolonged oscillation of the beam.

Theoretical equations describing the behavior of the elastic systems employed in these balances have not been published. Equations of this kind are, however, highly useful, both in understanding the behavior of elastic balances and in solving problems of design and construction. The author has, accordingly, derived expressions relating the sensitivity of the cantilever, Kirk-Craig-Gullberg, and Garner balances to structural and load parameters. The formulas presented hold accurately for small deflections only, but may be used as rather close approximations for almost all cases of practical interest.

Before presenting the equations, it is desirable to make a distinction between the "sensitivity" and the "sensibility" of a balance. "Sensitivity" shall be defined here as the magnitude of the response of the elastic system per unit of mass of load. (In some cases it is more convenient to express the response in terms of linear displacement, and in others in terms of angular displacement.) "Sensibility" shall be defined as the minimum mass which may be detected with the balance. In general, "sensibility" will have a statistical rather than an absolute significance and will represent the threshold of detection of response. The sensitivity of a particular elastic system is fixed by specifying its geometry and load, but the "sensibility" of a balance employing this system will depend upon the method used to detect or measure the response.

If the minimum observable response is ρ_{min}, then the sensibility, M_{min}, will be related to the sensitivity S by the equation:

$$M_{min}\ (\text{gm}) = \frac{\rho_{min}\ (\text{cm or radians})}{S\left(\frac{\text{cm or radians}}{\text{gm}}\right)} \qquad (1)$$

Cantilever balance. For a cantilever balance composed of quartz fibers of uniform circular cross section, as shown in the lower sketch in Fig. 1, sensitivity can be derived from Eq. 2 at the bottom of this page. In Eq. 2, M is the load in grams, g is the gravitational constant, h is the vertical distance* from the axis of the beam to the point of attachment of the suspension fiber, L is the distance from the fixed end of the beam to the junction with the suspension fiber, P is the length of the pointer, E_b and E_s are the values of Young's modulus for beam and suspension fibers, respectively, and r_b and r_s are their respective radii.†

It is evident from Eq. 2 that the sensitivity is dependent upon the load. If r_s is made much smaller than r_b and h is made very small, the change in sensitivity with change in load is negligibly small.

Equation 2 has been derived upon the assumption that the deflection of

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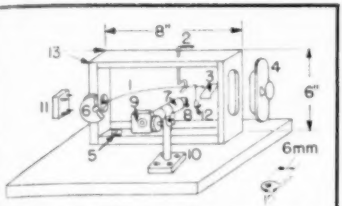
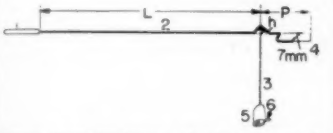
* In the equation as given, h is assigned a positive value if the point of attachment of the suspension fiber lies above the central axis of the beam, a negative value if it lies below.

† For small silica fibers, the value of Young's modulus varies appreciably with the diameter. Exact values may be found in chap. V of "Procedures in Experimental Physics," by J. Strong *et al.* (Prentice-Hall, Inc., New York, 1943).

$$S = \frac{gL^2}{3E_b\pi r_b^4}\left[\frac{4L\left(\frac{E_b\pi r_b^4}{4} + L\sqrt{\frac{E_s\pi r_s^4}{4Mg}} - hL\right) + \frac{3}{2}PE_b\pi r_b^4 - 3L^2\left(\sqrt{\frac{MgE_s\pi r_s^4}{4}} - hMg\right)}{\frac{E_b\pi r_b^4}{4} + L\sqrt{\frac{E_s\pi r_s^4}{4Mg}} - hL}\right] \qquad (2)$$



General Assembly: **1,** quartz fiber beam; **2,** beam arrest; **3,** aluminum reflector; **4,** door; **5,** ionizing source; **6,** wheel for adjusting angle of beam; **7,** brass tube; **8,** microscope objective; **9,** filar micrometer; **10,** metal stand; **11,** removable wood block; **12,** weighing pan of 0.5 mil platinum foil; **13,** thin glass plate.



Beam Construction: **1,** shank; **2,** beam; **3,** suspension fiber; **4,** pointer; **5,** pan hanger; **6,** pan

Dimensions are somewhat optional. Ordinarily, L and P are 12 and 3 cm respectively. The suspension fiber is 3 cm in length and 5 μ in diameter. The pan hanger is constructed from 75 μ diameter fiber. The platinum pan is 5 mm in diameter and 0.5 mil thick. For 0.01 μg sensibility the beam diameter is about 0.1 mm. The pointer is offset 7 mm to bring it nearer the glass side of the case. Except for the pan the parts shown are constructed of fused quartz.

FIG. 1. Simple cantilever balance

the beam is very small. The deflection due to the weight of the beam itself has been neglected. For fibers of very small diameter, the self deflection becomes large and imposes a practical limit on the sensitivity that can be obtained with the cantilever-type balance.

The deflection at the free end of a straight horizontal fiber of uniform cross section may be computed approximately from the following equation:

$$\text{self deflection} \cong \frac{M_b g L^3}{2E_s \pi r_b^4} = \frac{\delta g L^4}{2E_s r_b^2} \quad (3)$$

where M_b is the mass of the beam, δ is the density of the beam material and the other symbols have their previous significance. For a beam of given length, the self deflection is proportional to the reciprocal of the square of its radius. Because of the self deflection of the beam, the sensitivity of a quartz cantilever balance cannot be made much greater than 10^4 cm per gm.

Simple torsion balance. For a simple torsion balance of the design shown in Fig. 2, the sensitivity equation is:

$$S\left(\frac{\text{rad}}{\text{gm}}\right) = \frac{gL}{\frac{\eta \pi r_t^4}{2T} - M_b d + 2Mg\left(\sqrt{\frac{E_s \pi r_s^4}{4Mg} - h}\right)} \quad (4)$$

where g is the gravitational constant, L is the length of one arm of the beam,* η is the shear modulus for the torsion fiber, r_t is the radius of the torsion fiber and T its length, M_b is the mass of the beam, d is the distance from the center of mass of the beam to its center of rotation, M is the mass suspended from each arm, E_s is Young's modulus for the suspension fibers and r_s their radii, and h is the normal distance from the line drawn through the points of attachment of the suspension fibers to the parallel plane passing through the axis of rotation.

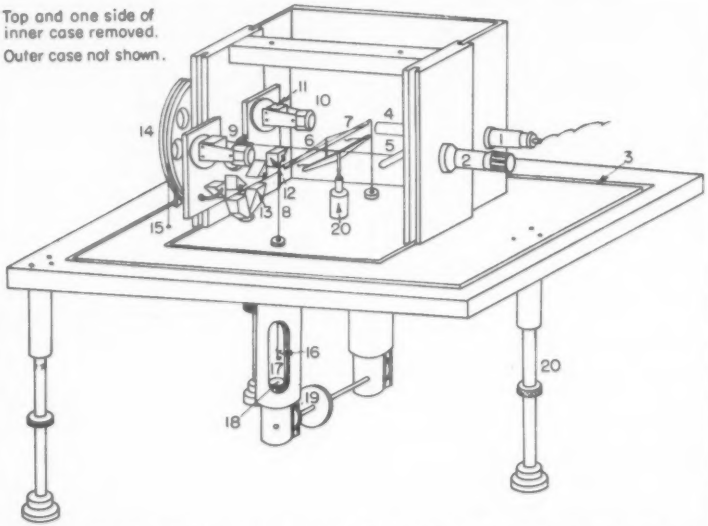
Both d and h may have negative, as well as positive, values. Equation 4 holds if negative values are used when the corresponding points lie below the center of rotation of the system.

If d and h have zero or negative values, the balance is stable at all loads. If d is positive and h is zero, the balance is stable at all loads for all values of d less than:

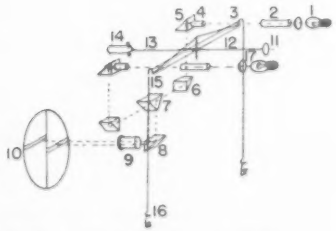
$$\frac{\eta \pi r_t^4}{2M_b T} \quad (5)$$

If $d > (\eta \pi r_t^4)/(2M_b T)$, the balance

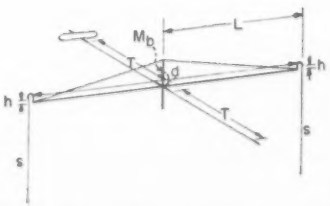
* The beam is assumed to be symmetrical about the axis of rotation and to be supported by two identical fibers of length T.



General Construction: **1,** light source; **2,** torsion fiber tension; **3,** groove for outer case; **4,** lucite rod; **5,** quartz bow; **6,** torsion fiber; **7,** beam; **8,** hang-down fiber; **9,** collet attached to axle of torsion wheel; **10,** low-power objective; **11,** pair of right-angle prisms; **12,** right-angle prism; **13,** prism to produce juxtaposition of fields; **14,** torsion wheel; **15,** cord for turning torsion wheel; **16,** pan well; **17,** pan hanger; **18,** pan arrest; **19,** pan arrest control wheel; **20,** adjustable leg



Elastic and Optical Systems: **1,** light source; **2,** lucite rod; **3,** index fiber; **4,** low power objective; **5,6,5,** right angle prisms; **7,** silvered right angle prism; **9,** ocular; **10,** ground glass screen; **11,** quartz bow; **12,** beam; **13,** torsion fiber; **14,** torsion wheel axle; **15,** hang-down fiber; **16,** pan hanger; **17,** condensing lens. (*NOTE:* The optical system shown is that employed in a modified form of the Kirk-Craig-Gullberg balance designed by the author, and differs somewhat from the system shown above.)



Detail of Beam: Dimensions are somewhat optional, but in most models $L = 5$ cm, $T = 5$ cm length, 24.5 μ diameter. Except for the 75 μ diameter diagonal fibers, the beam is made from 200 μ fibers. The suspension fibers and index fiber are 5 μ in diameter. Center of mass, M, is very near the center of rotation. Normally, h is made as small as possible. All parts shown are made of fused quartz.

FIG. 2. Kirk-Craig-Gullberg quartz microgram balance

will be unstable at all loads less than

$$M = \frac{\left(M_b d - \frac{\eta \pi r_f^4}{2T}\right)^2}{g E_s \pi r_s^4} \qquad (6)$$

but will become stable above this minimum load with subsequent decreasing sensitivity with increasing load.

If h is positive and d is zero, the balance will be unstable at all loads greater than:

$$M = \left[\frac{\sqrt{g E_s \pi r_s^4} + \sqrt{g E_s \pi r_s^4 + \frac{4gh\eta\pi r_f^4}{T}}}{4gh}\right] \qquad (7)$$

Below this value the sensitivity will diminish as the load is diminished.

If both d and h are positive and $d > (\eta \pi r_f^4)/(2M_b T)$, the balance will be unstable except with a range of load. See Eq. 8 at the bottom of the page.

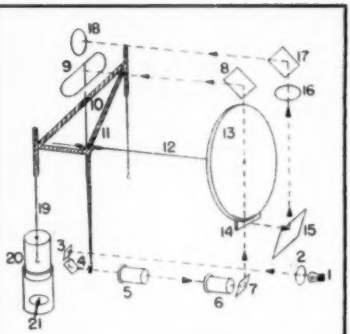
Within this range the balance will be stable, with varying sensitivity, which will approach infinity as the load approaches the maximum or minimum value. At intermediate loads the sensitivity will be less, and at some particular value of M will exhibit a minimum. This occurs when

$$M = \frac{E_s \pi r_s^4}{16 g h^2} \qquad (9)$$

The minimum sensitivity will be:

$$S_{min} = \frac{gL}{\frac{\eta \pi r_f^4}{2T} - M_b d + \frac{E_s \pi r_s^4}{8h}} \qquad (10)$$

Torsion balance with vertical suspension. In the case of the torsion balance with vertical suspension, the vertical suspension fiber introduces an additional elastic element into the system.



1, light source; **2,** condensing lens; **3,4,** plane mirrors; **5,6,** 15 × microscope oculars; **7,8,** plane mirrors; **9,** ground screen; **10,** vertical suspension fiber; **11,** beam; **12,** torsion fiber; **13,** torsion wheel; **14,** vernier; **15,** plane mirror; **16,** lens; **17,** plane mirror; **18,** lens; **19,** hang-down fiber; **20,** pan well; **21,** pan arrest

FIG. 3. Garner high-capacity quartz fiber balance

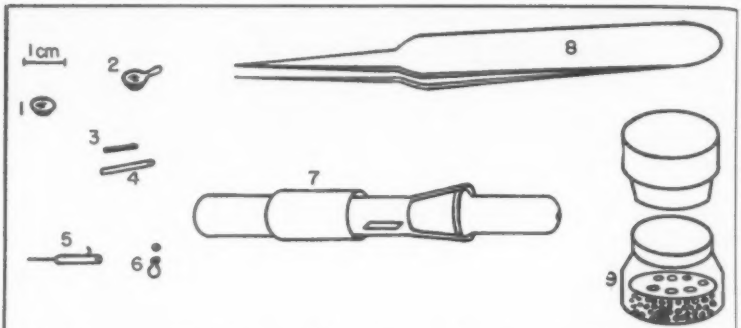
The sensitivity equation (Eq. 11), where E_v and r_v are Young's modulus and the radius respectively of the vertical suspension fiber, is given at the bottom of this page.

The Garner (*12*) high-capacity quartz microbalance (Fig. 3) is an example of a balance with a vertical suspension fiber. The Garner balance is so constructed that both d and h have negative values. It therefore is unstable with loads < 0.6 gm and > 5 gm.

From Eqs. 4 and 11 it is evident that either a simple torsion balance or torsion

$$M = \left[\frac{\sqrt{g E_s \pi r_s^4} \pm \sqrt{g E_s \pi r_s^4 - 8gh\left(M_b d - \frac{\eta \pi r_f^4}{2T}\right)}}{4gh}\right] \qquad (8)$$

$$S = \frac{gL}{\frac{\eta \pi r_f^4}{2T} - M_b d + 2Mg\left(\sqrt{\frac{E_s \pi r_s^4}{4Mg}} - h\right) + \frac{\sqrt{(M_b + 2M)g}\sqrt{E_v \pi r_v^4}}{2}} \qquad (11)$$



1,2, weighing pans of thin platinum foil; **3,4,** platinum and aluminum weighing tubes; **5,** glass or quartz weighing tube; **6,** glass weighing bottle with bead stopper; **7,** quartz micromuffle with platinum outer shield; **8,** jewelers forceps; **9,** micro dessicator

FIG. 4. Auxiliary apparatus used for microweighings

balance with vertical suspension may be constructed in such a way as to possess both high load capacity and high sensitivity. The elastic resistance of heavy supporting fibers may be largely counterbalanced by making d and/or h sufficiently positive.

In the case of a simple torsion balance of high capacity designed for use as a null instrument, the heavy supporting fibers obviously cannot be used as torsion fibers. The design must be altered so that a separate torsion fiber is used to apply the torque necessary to restore the balance to the null position.

It should be emphasized, of course, that for any balance used as a null instrument the sensibility of the balance imposes a limit upon, but does not necessarily determine, the precision of weighing. The precision may depend rather, upon the precision of measuring the force required to restore the balance to null position.

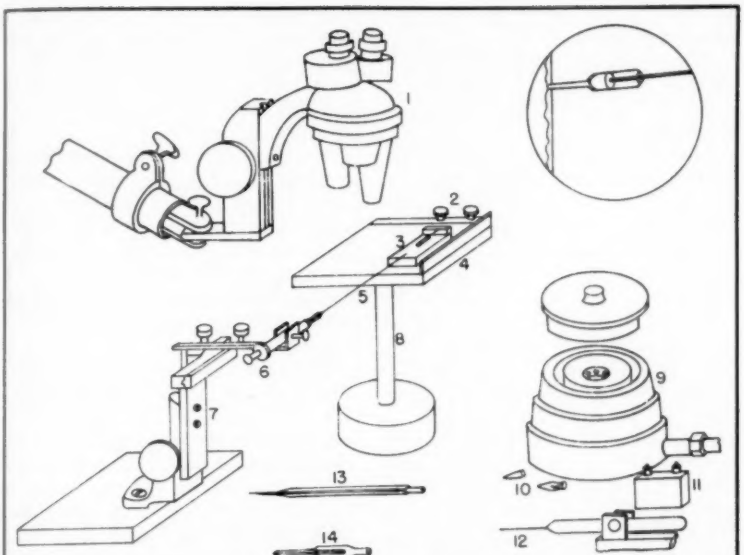
Finally, a few practical points connected with the use of highly sensitive elastic balances may be mentioned.

In general, these balances are less subject to environmental disturbances than are conventional microbalances. The cantilever and simple torsion balances of the types described above operate satisfactorily in nonthermostated rooms. Although they are somewhat sensitive to vibrational disturbances, the vibrations of the balance damp out very quickly. Special mountings are not required unless the balances are exposed to continuous and rather strong vibrations.

Static electrical effects may sometimes disturb the weighings, especially in very dry weather. It is common practice to maintain an ionizing source inside the balance case. Alpha activity equivalent to about 10^8 counts per minute is satisfactory for this purpose.

The balances show no detectable change in sensitivity with use over long periods of time. A balance of the Kirk-Craig-Gullberg design, in use in the author's laboratory for three years, has shown no detectable change in sensitivity ($< 0.1\,\%$).

Although these balances are obviously fragile instruments, they are not subject to excessive breakage, even when used by relatively untrained personnel. The balance referred to above has been broken but once during the period mentioned, although eight different



1, microscope; 2, mechanical stage; 3, metal slide; 4, microcone; 5, micropipet; 6, syringe control; 7, micromanipulator; 8, metal stand; 9, air driven centrifuge; 10, microcones; 11, magnetic buzzer; 12, glass stirring thread; 13, graduated pipet; 14, self-filling pipet

FIG. 5. Apparatus for microgram scale solution chemistry

persons have been trained in its use during this tine. The cantilever-type balances have been found to be satisfactory for class use in a course in microchemical techniques for college seniors.

Auxiliary apparatus used in weighings with the sensitive elastic balances is shown in Fig. 4. Most of the pieces shown are manipulated with the aid of jeweler's forceps. Usually, the balances are loaded by hand. Occasionally they may be loaded with the aid of a simple manipulator, such as that shown in Fig. 5.

Solution Chemistry-Microgram Scale

Apparatus and general techniques. The apparatus illustrated in Fig. 5 includes most of the pieces ordinarily used in working with aqueous solutions on the microgram scale. Equipment of this kind was found suitable for the concentration, purification and isolation of the first pure samples of plutonium (*2*), neptunium (*13*), americium (*14*), and curium (*15*).

The microscope and stage assembly are somewhat similar to the apparatus described by Benedetti-Pichler (*3*) for qualitative analysis on the microgram scale. At ordinary operating magnifications, however, the optical system provided by a wide field stereoscopic microscope is distinctly superior to that afforded by the standard binocular microscope recommended by Benedetti-Pichler. The microscope mounting and stage assembly have been modified to give greater flexibility and ease of operation. Improved precision in the measurement of reagents has been

attained by the use of carefully calibrated micropipets.*

The preparation and analysis of thorium oxalate may be described in order to illustrate the techniques employed:

Five microliters of an approximately 0.003*M* solution of thorium nitrate in 1*M* HNO_3 is withdrawn from a prepared stock solution, using a 1-ml syringe control and a pipet of the type marked 13 in Fig. 5. The pipet is then mounted in the clamp of a simple manipulator as shown in Fig. 5.† A 20-μl quartz cone (numbered 5 in Fig. 4) previously weighed to the nearest 0.01 μg is clamped to a metal slide which is then mounted on a mechanical stage. The cone is brought into the field of view of the microscope. The tip of the pipet is advanced to within a millimeter of the open end of the cone and carefully aligned to coincide with the central axis of the cone. Final adjustment is made by viewing the pieces through the microscope. The tip of the pipet is then moved inward until it is in contact with the bottom of the cone.

The plunger of the control is advanced slowly by a spiral movement until some liquid is forced into the cone. Delivery is continued at a slow constant rate, the tip being gradually withdrawn as the liquid enters the cone. In this way any excessive wetting of the outside of the pipet tip is avoided. (In some cases it is advantageous to coat the tip of the pipet with a thin film of paraffin, so that it is not wet by the solution.) Complete emptying of the pipet should not be attempted, since the sudden change in pressure releases a large volume of air and the solution may spatter on the walls of the cone. All but about 1% of the solution may be safely expelled, however, and a single rinse insures essentially quantitative transfer of the solute.

The pipet is withdrawn and the tip touched to a 2–3 μl droplet of 0.5*M* HNO_3 placed on a paraffin-coated slide. The entire droplet is withdrawn into the pipet. The pipet is then filled to the mark with water from a second droplet and again placed in the manipulator. The rinse solution is added to the cone.

Two μl of 0.6*M* oxalic acid are added from a second calibrated pipet and the solution is stirred by means of a magnetically activated glass stirring thread, and allowed to stand for one hour. The mouth of the cone is covered with "parafilm"‡ during this time. The covered cone is placed in a larger tube, also sealed with "parafilm" and centrifuged in a small air-driven centrifuge§ (piece 9 of Fig. 5). The cone containing the oxalate is returned to the stage and the supernatant liquid withdrawn carefully into a pipet with a very fine tip. The oxalate is washed with three 2 μl portions of distilled water, and then dried by directing a stream of warm, filtered air into the cone. The tube is weighed to ± 0.01 μg on a suitable balance.

The oxalate is then dissolved in 2*M* sulfuric acid solution, transferred with rinsing to a small flat porcelain dish, treated with an excess of standard 0.005*M* ceric sulfate solution, and back titrated with ferrous ammonium sulfate with a ferrous phenanthroline indicator. A blank titration is performed to correct for the end-point error. The type of buret used is that described by Sisco,

* Pipets of this type are calibrated for content by weighing the quantity of mercury delivered by the pipet. A precision of ± 0.2% is readily obtainable.

† Manipulators of this design are available from the Gamma Scientific Co., Great Neck, N. Y.

‡ A semi-plastic tough paraffin-like tissue manufactured by the Menasha Products Co., Menasha, Wis.

§ Available from Microchemical Specialties Co., Berkeley, Calif.

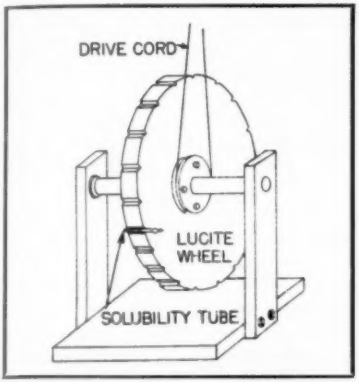


FIG. 6. Microsolubility apparatus. Solubility tubes are conveniently held in place by a rubber band around the rim of the wheel

Cunningham and Kirk (*16*). General titration procedures on this scale have been developed by Kirk and coworkers (*5*). The standard error of the titration is about ±0.3%.

Thorium is determined on a sample similarly prepared, by igniting the oxalate to the oxide in a small platinum dish. The oxide is weighed on a quartz balance.

Obviously, a large number of common analytical operations may be carried out by techniques similar to those outlined above.

Solubility measurements. Accurate measurements of the solubilities of various compounds of the synthetic elements have been obtained on very small samples, as little as a few hundredths of a microgram in some cases (*2*). Such measurements are facilitated by radiometric assay of the solution in equilibrium with the solid phase. In the case of Pu^{239}, for example, as little as 0.01 μg may be determined with an error of less than 2%.

Solubility measurements on nonradioactive materials require a minimum of several micrograms of sample since, in general, precise chemical analysis cannot be carried out with smaller samples.

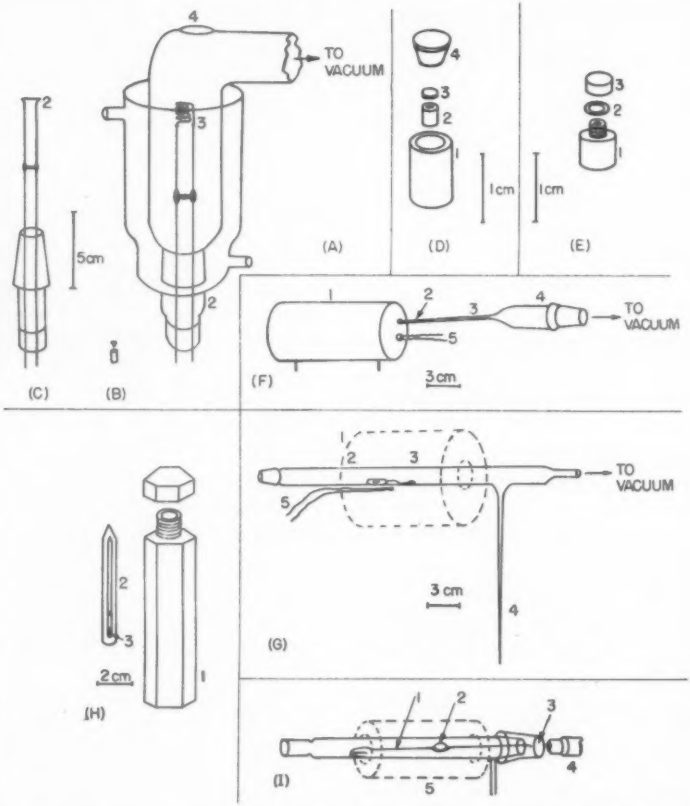
Apparatus designed by Stewart and Cunningham (*17*) for solubility measurements on very small samples is illustrated in Fig. 6. Solid and solvent are placed in glass capillaries which are then sealed and attached to the periphery of a notched wheel that is rotated by means of a motor. Rotation of the wheel produces a continuous mixing of the solid and liquid phases. The wheel may be mounted in an air or water thermostat if desired. Upon completion of a run, the solid phase is centrifuged to the bottom of the tube, which is then broken open so that samples of the supernatant liquid can be withdrawn into calibrated pipets for analysis. If desired, the tube may be resealed and the run continued.

The samples of supernatant solution are spread on thin plates and dried for radiometric assay, or are used for some other type of chemical analysis. In the case of the rare-earth elements, for example, some residues may be converted directly to oxides by ignition. The oxides may then be weighed on a quartz balance.

If the composition of the solid phase is unknown, it may be established in some cases by X-ray diffraction methods which require but a few micrograms of sample (see section on *X-ray diffraction*, page 78).

Specific activity and half-life determinations. The decay constants and half-lives of several long-lived alpha-emitting isotopes (Pu^{239}, Np^{237}, Am^{241}) have been accurately determined by weighing microgram samples of the pure isotopes and measuring the radioactivity associated with the weighed samples.

Precise measurements of this kind require that the chemical and isotopic composition of the sample weighed be accurately known, and that the counting yield of the instrument used to



(A) **Westrum Vacuum Furnace**: **1**, 80-mil wolfram rod; **2**, water-cooled joint; **3**, heating coil **4**, window

(B) **Refractory Crucible**

(C) **Filament-type Heater**: **1**, wolfram rod; **2**, wolfram or tantalum ribbon

(D) **Double Crucible System for Metal Production**: **1**, outer crucible of BeO, ThO_2, TaC, *etc.*; **2**, inner crucible; **3**, inner crucible lid; **4**, outer crucible plug

(E) **Platinum Weighing Bottle**: **1**, body; **2**, teflon washer; **3**, screw cap

(F) **Fried's Halide Apparatus**: **1**, furnace; **2**, halide sublimate; **3**, quartz capillary; **4**, quartz joint; **5**, thermocouple

(G) **Lohr, Broido, and Cunningham Halide Apparatus**: **1**, furnace; **2**, magnetic pusher; **3**, sample boat; **4**, capillary; **5**, thermocouple

(H) **Microbomb**: **1**, steel casing; **2**, quartz capillary; **3**, sample

(I) **Broido and Cunningham Gravimetric Apparatus for Solid-gas Reactions**: **1**, cantilever balance; **2**, pan for sample; **3**, flat window; **4**, microscope; **5**, furnace

FIG. 7. Apparatus commonly used in microwork with solid compounds

measure the activity also be known.

In practice, isotopic purity may be insured, either by selection of the proper method of preparing the isotope or by reference to some method of isotopic analysis such as that afforded by the mass spectrograph or alpha pulse analyzer in the case of alpha-particle emitters.

Chemical purity may be insured by extreme care in preparing the material to be weighed, by reference to spectrographic analysis* of a portion of the prepared material and by repeated measurements of the specific activity following additional cycles of chemical purification.

If the isotope is weighed in the form of its oxide (as is often convenient) it may be difficult to determine the precise composition of the oxide. X-ray diffraction measurements establish the type of structure, but not necessarily the exact composition, since an appreciable fraction of the lattice sites may be vacant. This difficulty may be avoided if the isotope can be prepared and weighed in the form of metal.

The precise determination of the radioactivity associated with the sample or a known aliquot of it requires that the geometry of the counter used be known, that counting losses due to coincidence and self-absorption be corrected for, that an appropriate background correction be made, and that backscattering be taken into account.

In practice, low geometry counters (geometry $< 1\%$) are used to avoid corrections due to low-angle scattering of alpha particles and to minimize losses due to self-absorption of the sample. Background and coincidence corrections are carefully established. The samples are mounted as thin, uniformly spread layers on smooth flat plates of nonabsorbent material (such as platinum or quartz; the latter has the advantage that low-angle scattering of alpha particles by the quartz is almost negligible). Generally, "weightless" samples are prepared for counting by dissolving the weighed sample, diluting to a known volume with residue-free reagents and mounting a very small aliquot on a plate of platinum or quartz. Uniform distribution of the sample on the plate is aided by careful cleaning of the plates before adding the sample, or by the use of organic spreading agents. The samples are then dried and ignited to a dull red heat to dry them and to burn off organic matter.

Chemistry of Solid Compounds

Apparatus and general techniques. Much of the investigation of the chemical properties of the actinide and other rare elements has been carried out on solid compounds.

Success in handling very small samples of labile solid compounds requires extremes of precaution well beyond that which is adequate for successful handling of macro amounts. For example, a few micrograms of water vapor adsorbed on the walls of a tube are more than sufficient to convert a 10-μg sample of samarium trichloride to the oxychloride, whereas the

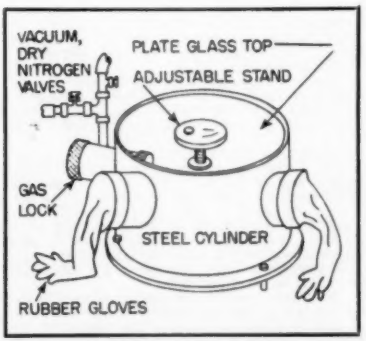


FIG. 8. Dry box

* Spectrographic analysis employing copper electrodes and the spark method will detect most common impurities at a level of 0.1 μg or less.

same amount of water can scarcely affect the composition of a gram of the same compound.

There are three general requirements for successful work with small quantities of solid compounds:

1. a system capable of producing and maintaining reasonably high vacuums (10^{-6}–10^{-7} mm Hg),
2. extreme care in outgassing the apparatus used,
3. careful purification of gaseous reagents introduced into the apparatus.*

The types of apparatus most frequently used in investigating small quantities of solid compounds are illustrated in Fig. 7. The various uses of these pieces of equipment is discussed in the succeeding section.

Methods of preparation and investigation of various types of compounds:

Oxides. The common oxides of the actinide and rare-earth elements may be prepared by igniting in air a few micrograms of the oxalate, hydroxide, peroxide, nitrate, *etc.* on a thin platinum foil, by means of a furnace of the type marked 7 in Fig. 4. The resulting product may then be transferred to a capillary tube for examination by X-ray diffraction or other methods.

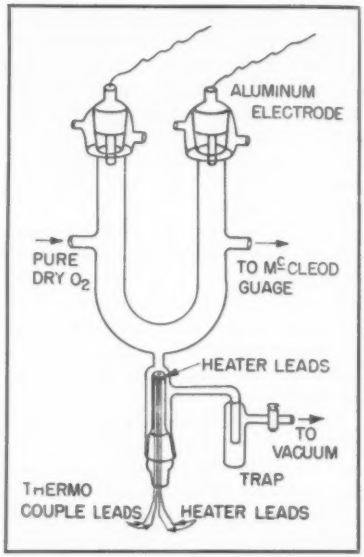


FIG. 9. Gruen and Katz atomic oxygen apparatus

The transfer of the prepared oxide is effected by using a fine wolfram needle† to pick up individual pieces of the oxide deposit. The transfer is carried out under about 15–30 times magnification, using a stereoscopic microscope. Hygroscopic or radioactive samples are handled inside a closed chamber such as that in Fig. 8. The design shown is due to Westrum (*19*).

Oxide intended for weighing as part of a specific activity determination is prepared in place on a thin (0.5 mil) platinum weighing pans.

If the oxide is to be used as a starting material for preparing other compounds (halides, for example), it is prepared in finely divided form by low-temperature ignition of oxlaate or hydrous oxide, rather than by decomposition of nitrate. High temperatures

* Very satisfactory methods of dehydrating and purifying several common gases have been developed by Newton (*18*). Very pure dry hydrogen may be produced by the thermal decomposition of uranium hydride. The hydride is made initially from uranium metal and impure hydrogen.

Helium, argon and other noble gases may be dried and rendered free of oxygen, nitrogen, CO_2, NH_3 and hydrocarbon gases by passing them over uranium metal turnings at 750°–800° C.

Hydrogen chloride and hydrogen bromide may be almost completely freed of water vapor, oxygen and elementary halogens by passing them over the respective anhydrous uranium halides at 400° C. The uranium halides are formed in place in the drying tube by reaction of uranium metal with the hydrogen halide. (Allowance must be made for large changes in bulk density which occur when the metal is converted to the trihalide.)

Nitrogen may be purified from oxygen by passing the gas over uranium nitride at 600° C.

The original paper should be consulted for technical details concerning these methods of purification.

† These needles may be sharpened by stroking the hot metal with a lump of sodium nitrite.

(> 600° C) are avoided, since they produce a less reactive form of the oxide.

Since most of the synthetic elements exhibit variable valence, more than one oxide is usually known for a particular element.

Oxides of higher average oxidation number than that obtained by air ignition have been prepared by various methods.

Gruen and Katz (*20*) have described apparatus suitable for the small scale preparation of higher oxides by treatment of the lower oxides with either NO or atomic oxygen. The type of apparatus used by them is illustrated in Fig. 9.

Higher oxides have also been prepared by bomb methods and high pressures (100–200 atmospheres) of oxygen. The method was used originally by Fried and Davidson (*21*) for the attempted preparation of a higher neptunium oxide, and has been used recently by Eyring (*22*) for the preparation of PrO_2. The type of bomb used is depicted in Fig. 7.

Lower oxides have been prepared in most cases by reduction with hydrogen, using apparatus similar to that marked *A* in Fig. 7. The starting oxide is contained in a crucible of platinum, tantalum or other suitable material. The stoichiometry of the reduction may be determined by using a platinum vessel of the design shown by *E* in Fig. 7 (*22*). Upon completion of the reduction, the hydrogen is pumped off at elevated temperatures, and the system cooled and flushed with pure dry argon. Argon is kept flowing while the vessel containing the sample is removed from the apparatus and quickly capped. The vessel is then weighed on an ordinary microbalance.

A lower oxide of americium has been prepared by thermal decomposition of the higher oxide in a platinum vessel at high temperature in high vacuum (*23*).

Equilibrium investigations of the thermal decomposition of praseodymium oxides have been carried out by Asprey (*24*) with samples of only a few milligrams weight. The apparatus employed is illustrated in Fig. 10. The general reliability of the apparatus and techniques employed was first tested

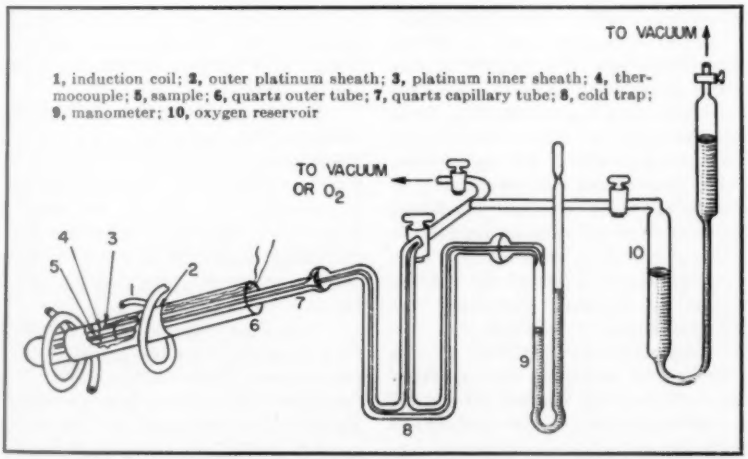


FIG. 10. Asprey and Cunningham oxide decomposition apparatus

by investigating the thermal decomposition of CuO. Data obtained on a 10-mg sample checked the very accurate ΔH value for the reaction: $CuO \rightarrow \frac{1}{2}Cu_2O + \frac{1}{4}O_2$ reported by Randall, Nielson, and West (*25*) to within ±0.2%.

Halides. Chlorides, bromides, and iodides of the actinide elements have been prepared by several techniques suitable for very small amounts.

The apparatuses most frequently used for this purpose are illustrated by *F* and *G* in Fig. 7. The design of *F* in Fig. 7 is due to Fried and Davidson (*21*). A few micrograms of oxide are introduced into the thin-walled quartz capillary tube, which is then attached to the vacuum apparatus. The furnace is put in place, the system is evacuated, and the halogenating agent admitted to the capillary which has been heated previously to an appropriate temperature.

Fried has used CCl_4 for the preparation of chlorides from oxides, and $AlCl_3$, $AlBr_3$ and AlI_3 for the preparation of chlorides, bromides, and iodides from oxides. In the latter case, the aluminum halide is sublimed in substantial quantity onto the oxide, the capillary is sealed, enclosed in a jacket of heavier tubing, and heated at 500° C for several hours. The capillary is then sealed with apiezon wax into a larger tube in such a way that the tip projects into the bore of an open stopcock which communicates with a vacuum system. When the stopcock is turned, the tip of the capillary is broken. Fractional sublimation of the reaction products is then carried out in vacuum.

At the completion of the run the section of capillary containing the halide sublimate is sealed off.

Oxyhalides. Oxyhalides of the actinide and rare-earth elements have been prepared by thermal decomposition of the hydrated halides and by the vapor-phase hydrolysis of the anhydrous halides in a mixture of H_2O and HCl vapor. The latter method is suitable for an investigation of the equilibria involved in reactions of this type. Two types of apparatus employed by Broido and Cunningham (*26*) in an investigation of the vapor-phase hydrolysis of rare-earth trichlorides are shown in Fig. 7. Samples of the product may be removed from the system without interruption of the gas flow, using the apparatus marked *G* in Fig. 7. The progress of the reaction between a solid and gas may be followed quantitatively with the apparatus shown by *I* in Fig. 7.

Sulfides, nitrides, carbides and silicides. Sulfides of several of the actinide elements have been prepared by treating oxides or halides with H_2S or mixtures of H_2S and CS_2 in quartz tubes at elevated temperatures by means of apparatus somewhat similar to that shown by *E* in Fig. 7 (*21*).

Plutonium nitride has been prepared by treating the trichloride in a platinum vessel with NH_3 gas at elevated temperatures, or on a still smaller scale by heating pieces of metal on a platinum filament in the presence of anhydrous ammonia (*27*).

Plutonium silicide has been prepared in micro amounts by heating a mixture of the trifluoride with calcium silicide in a system similar to *A* in Fig. 7 (*28*).

Hydrides. Hydrides of various actinide metals have been prepared and investigated in an apparatus shown in Fig. 11. The design shown is similar to that devised originally by Baumbach (*29*). Hydride formation has frequently been used as a test to distinguish true metals from metallic looking lower oxides.

The apparatus shown is suitable for a quantitative investigation of the pressure-temperature-composition relationships in the metal-hydrogen system. Hydride is formed at a suitable temperature, with the mercury droplet confined to the depression in the glass

elbow. The joint is then rotated, plugging the capillary above the sample with the mercury droplet. The sample is then heated. When the pressure of hydrogen above the sample exceeds the pressure of hydrogen above the droplet, the mercury moves upward.

Metals. The preparation and investigation of the synthetic and other rare elements in metallic form have been an important part of the general study of the properties of these elements. A method for microgram-scale preparation of the actinide metals was devised originally by Baumbach and Kirk (*30*) and has been improved by Westrum and others. The improved method has been described by Fried and Davidson (*21*). The apparatus employed is shown by *A* and *B* in Fig. 7.

A charge of suitable compound (halide, for example) is placed in the bottom of a small, previously outgassed, inner crucible which is then closed with a loose-fitting cover. The inner crucible is inserted into the larger, outer crucible (also previously outgassed), a small piece of reductant metal is added, and the outer crucible is closed by a channeled plug. The assembly is supported in the interior of a wolfram heating coil, which is surrounded by a radiation shield.

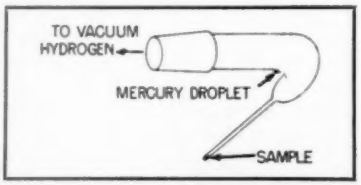


FIG. 11. Baumbach hydride apparatus

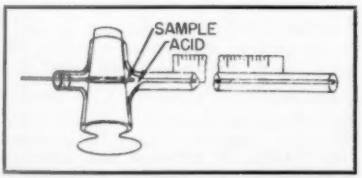


FIG. 12. Hydrogen evolution apparatus

Shield, coil, and crucible assembly are inserted into a vacuum line as shown, and pumped down to a pressure of 10^{-5}–10^{-6} mm Hg. The crucible is heated electrically to a suitable temperature, volatilizing the reductant metal, the vapor of which reacts with the halide to yield metal and reductant halide. The slag formed is largely absorbed in the refractory material. If reduction is carried out above the melting point of the product metal, the entire yield will usually be obtained in the form of a very few pieces.

The metal is removed from the crucible in a dry box and is freed of surface contamination with suitable needles and scrapers. These operations are carried out under a low-power microscope. If the crucibles are carefully prepared, inclusions of refractory material in the metal are rare. The clean metal is mounted in "lucite" or similar material for grinding and polishing and examination by the usual methods of metallurgical microscopy. Alternatively it may be used for measurements of the heat of solution, for observations of the melting point, determination of density, *etc.*

Various tests may be applied to determine the purity of the metal prepared by the method outlined above.

Hydrogen evolution from a weighed sample dissolved in dilute acid may be used as a method of assay. Micro apparatus suitable for this purpose is shown in Fig. 12 (*31*). The method is subject to a number of errors. The solution in the apparatus must be saturated with hydrogen prior to a run. Local depletion of hydrogen ion may produce hydrolysis and coating of the metal so that the reaction is incomplete. Appropriate corrections must be applied for the vapor tension of the solution and to obtain accurate measurements on microsamples, for the

radius of curvature of the bubble of hydrogen. The sensitivity of the method depends upon the difference in hydrogen equivalent of the metal and that of the impurities present.

Reversible hydride formation is frequently used to distinguish between poorly agglomerated metal and metallic appearing lower oxides.

Spectrographic analysis of the samples of 10-50 μg weight is suitable for the detection of many impurities.

Various Physical Methods and Measurements

X-ray diffraction. X-ray diffraction patterns by the powder method may often be obtained with a few micrograms of material. Compounds of relatively simple structure yield fewer and more intense lines, and in favorable cases have been identified at the level of less than 1 μg. The background produced by scattering of the X-rays by the material in which the sample is mounted is the most common factor limiting the production of an observable pattern.

The electromagnetic radiation associated with radioactive materials causes darkening of the X-ray film and, in some instances, severely limits the quantity of material that may be used for X-ray diffraction.

The preparation of quartz (or pyrex) capillary tubes suitable for X-ray diffraction investigation of various compounds of the synthetic elements has been discussed by Fried and Davidson (*21*). The prepared capillary should have an inside diameter of not more than about 150 μ and a wall thickness not greater than about 10 μ. The end is sealed off in a small flame. Excessive thickening of the quartz at the sealed end must be avoided.

Nonreactive samples may be mounted in a droplet of "Duco" or other organic cement stuck on the end of glass fiber.

In the absence of other information, diffraction patterns obtained by the powder method are not unequivocal indications of chemical composition. The presence of several percent of other phases frequently cannot be observed, compounds of quite different composition may yield identical structures and nearly the same lattice constants; complex structures are not readily identified, and random lattice deficiencies are not directly observable. Nevertheless, the method is a convenient and reliable one for distinguishing between reasonable alternative compositions, and very frequently enough additional information is available to make the identifica-

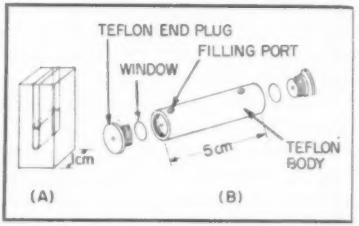
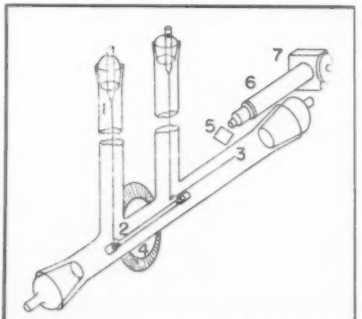
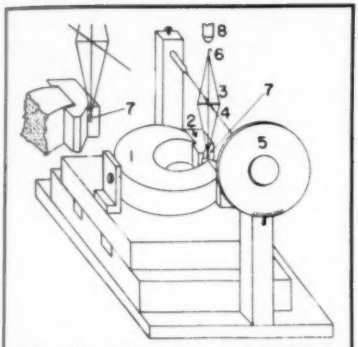


FIG. 13. Absorption cells. (A) Laminar type. (B) Kirk-Rosenfels-Hanahan type



1, 140-cm glass suspension fiber; **2**, thin-wall capillary with central partition; **3**, pointer; **4**, magnet pole face (only one shown); **5**, mirror; **6**, microscope; **7**, filar micrometer

FIG. 14. Apparatus used by Howland and Calvin to measure the magnetic susceptibility of aqueous ions of the actinide elements



1, 5-in. Alnico No. 5 magnet; **2**, specially cut pole piece to give uniform dH/dx; **3**, quartz frame; **4**, torsion fiber; **5**, torsion wheel; **6**, pointer; **7**, sample tube; **8**, microscope

FIG. 15. Cunningham and Wallman magnetic susceptibility apparatus for solids

tion virtually positive. An important advantage is that the sample is not destroyed by this method of analysis.

Absorption spectra. Absorption spectra may be obtained with most conventional apparatuses on much smaller samples than those customarily employed, by suitably altering the dimensions of the absorption cells. In most instruments, the incident light beam is much narrower than the cells usually employed. The use of narrower cells permits a corresponding reduction in the quantity of material required for determinating the spectrum.

Micro absorption cells, suitable for use with the Beckman model DU spectrophotometer are shown in Fig. 13.

The capillary-type cell is that devised by Kirk, Rosenfels, and Hanahan (*32*) (see *B* in Fig. 13) who have described quantitative methods of colorimetric analysis for a number of common elements on the milli-microgram (0.001 μg) scale. A special carriage* must be used with these cells, since they require very accurate alignment with the light beam from the spectrophotometer.

Cells of type *A*, Fig. 13, may be used with the carriage ordinarily supplied with the Beckman model DU. These cells are available commercially.† The quantity of material required to fill the cell is only about $\frac{1}{10}$ that necessary for the regular 1-cm cells.

The absorption spectra of solid compounds (actinide and lanthanide halides) have been obtained by Freed and Leitz (*33*) with samples weighing only a few micrograms, disposed on the walls of a thin quartz capillary tube. The method employed was to use a microscope to project a magnified image of microcrystals of the halide onto the slit of a prism spectrograph.

Jones and Conway (*34*) have used a microscope to project an image of rare-earth crystals onto the slit of a Baird 3-meter grating spectrograph, in order to investigate the fine structure of the absorption spectra of these compounds. With glass optics, the spectral range is limited to the region 3,600 A.U.–3 μ.

Magnetic susceptibilities. Methods have been devised for measuring the magnetic susceptibilities of very small amounts of substances of moderate susceptibility, or of moderate amounts of substances of low susceptibility. Three designs of apparatus are shown in Figs. 14, 15, and 16. The design shown in Fig. 14 is suitable for examining solutions or other liquids and was employed by Howland and Calvin (*35*) to measure the susceptibilities of various aqueous ions of the actinide elements, as an aid in determining their electronic structures.

The apparatuses shown in Figs. 15 and 16 were designed by Cunningham and Wallmann (*36*) for rapid room-temperature measurements of the susceptibilities of milligram samples of solid

* Obtainable from Microchemical Specialties Co., Berkeley, California.

† From Pyrocell Mfg. Co., New York, N. Y.

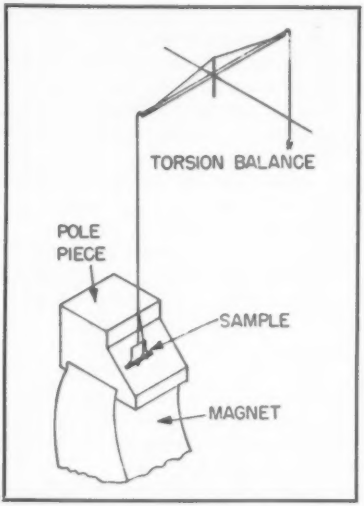


FIG. 16. Magnetic susceptibility measurement using a sensitive quartz torsion balance and an inhomogeneous field

compounds of the rare earths and actinide elements. The inhomogeneous field principle is employed. The apparatus is simple and capable of good accuracy.

Thermochemical measurements. Many of the developments in the field of microcalorimetry up to 1945 have been reviewed by Swietoslawski (*37*).

A semimicro and a microcalorimeter, especially suitable for thermochemical investigations of the synthetic elements, have been designed by Westrum (*38*). The microcalorimeter is of a semi-adiabatic type with exceptionally low heat capacity.

The principal features of the microcalorimeter chamber and surrounding jacket are indicated in Fig. 17. The instrument is sensitive to 10^{-4} cal. Accurate heats of solution of several actinide metals have been obtained with fractional milligram samples. With this instrument, it has been found possible to check the accurately

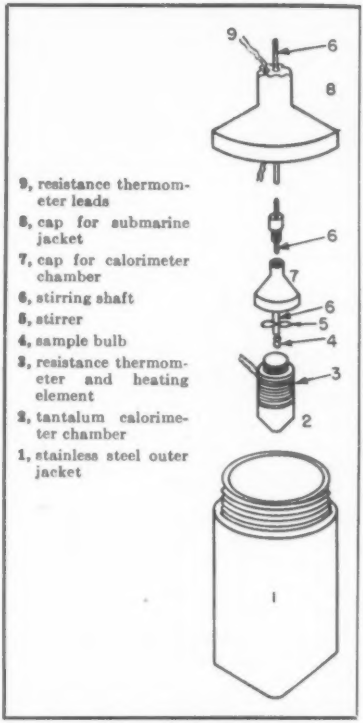


FIG. 17. Microcalorimeter chamber and submarine jacket

known heat of solution of magnesium metal to within $\pm 0.1\%$ with samples as small as 100 μg.

In common with most instruments of this type, precise measurements can be obtained only when the heat change being measured occurs rapidly, preferably in less than five minutes. However, the instrument has given fairly accurate results in measuring the heats of solution of samples of certain oxides, which require somewhat longer times to dissolve (7–10 minutes) (*22*).

Vapor pressure measurements. Vapor pressure measurements by the effusion technique have been carried out

with small quantities of various radioactive materials. The vapor is collected on plates suitable for direct radiometric assay of the vapor deposit. Apparatus suitable for vapor pressure measurements by this technique is shown in Fig. 18. The design is due to Westrum and Wallmann (*39*).

Extremely precise measurements of the vapor pressures of various plutonium halides have been obtained by Phipps, Simpson, and Seifert (*40*), with more elaborate apparatus.

Melting point. Various micro melting point apparatuses for moderately low melting substances are well known and need not be discussed. Most of the compounds of the synthetic and rare elements have relatively high melting points, however. Apparatus suitable for observing melting points up to about 1000° C is shown in Fig. 19.

One of the principal difficulties encountered in determining the melting points of high melting substances is that of providing a container or support that will not react with the sample at the high temperatures employed.

The melting points of small samples of reactive metals may be difficult to observe because of an oxide coating formed by reaction with oxygen or water desorbed from the sample support as the melting point is approached. The mechanical effect of the oxide coating in preventing observation of the melting has been overcome by Westrum and Eyring (*41*) by mounting the sample under slight tension between the jaws of a wolfram hairpin, as shown in the lower sketch in Fig. 19.

Density measurements. The densities of nonporous solids may be determined by a micro modification of the classical pycnometric method, using a capillary cell such as that shown in Fig. 20. The method was devised originally by Kirk (*42*). The design of the cell shown is due to Katz (*43*). The addition of a weighed amount of solid to the capillary tube produces a displacement of the liquid meniscus.

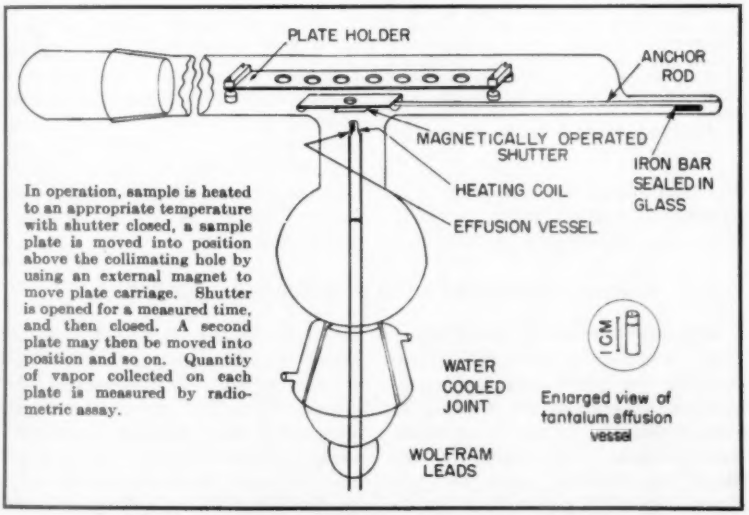
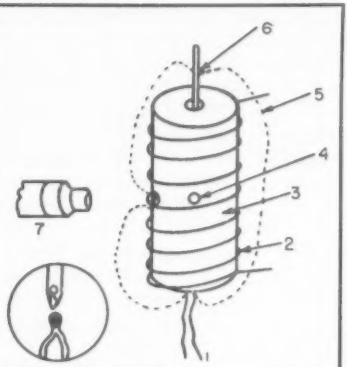
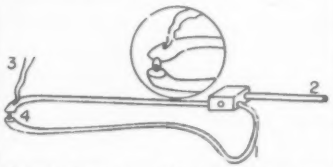


PLATE HOLDER
ANCHOR ROD
MAGNETICALLY OPERATED SHUTTER
IRON BAR SEALED IN GLASS
HEATING COIL
EFFUSION VESSEL
In operation, sample is heated to an appropriate temperature with shutter closed, a sample plate is moved into position above the collimating hole by using an external magnet to move plate carriage. Shutter is opened for a measured time, and then closed. A second plate may then be moved into position and so on. Quantity of vapor collected on each plate is measured by radiometric assay.
WATER COOLED JOINT
WOLFRAM LEADS
ICM
Enlarged view of tantalum effusion vessel

FIG. 18. Westrum and Wallman vapor pressure apparatus



Nickel Block Type: 1, thermocouple leads; **2,** resistance wire; **3,** nickel cylinder; **4,** illumination port; **5,** asbestos insulation; **6,** capillary containing sample; **7,** microscope. Inset shows enlarged view of capillary and thermocouple.



Hairpin Type: 1, wolfram hairpin; **2,** support rod; **3,** thermocouple; **4,** sample. Inset shows enlarged view of jaws with sample in place. Sample is heated by a surrounding coil of wolfram wire (not shown) and is observed with a low-power microscope.

FIG. 19. Melting point apparatus

The displacement is measured, with reference to a fiducial mark, by means of a filar micrometer. A few hundredths of a microliter of solid phase is sufficient for a volume measurement accurate to a few percent.

Protection of Personnel Handling High Level of Radioactivities

Most of the work with synthetic elements described above involves handling very high levels of alpha radioactivity. Such levels of radioactivity, particularly when in the form of finely divided solid materials, present a very serious physiological hazard. Proper protection of research personnel against this hazard is essential. The guiding principle, which has proved most satisfactory as a method of protection, has been to carry out all operations with such materials inside a closed system.

The general design of a box suited to micro work with high levels of alpha radioactivity is indicated in Fig. 21.

Where work must be done in more elaborate apparatus, a closed box of some light constructional material is erected around the equipment. The box is fitted with a suction hose equipped with filter and blower and vented in a suitable location, such as the flue of a fume hood. The suction maintains the pressure inside the box slightly below that of the surrounding atmosphere. Suitable windows and doors of "lucite" or other plastic are included in the box as needed. Holes admitting outside pipes, vacuum lines, *etc.*, are caulked with rubber or cellophane tape.

SEMIMICROMETHODS USED IN BOMBARDMENT WORK

Apparatus and General Techniques

It has become widely recognized in the last few years that semimicromethods of analysis are superior in several respects to the conventional macro methods. Their chief advantage lies in the improved speed and efficiency obtainable by use of the small-scale methods. Such methods have been used for several years in investigating nuclear reactions induced by artificial means. The advantages of increased speed in effecting chemical separations are particularly evident in work on spallation reactions at high energies where the product nuclei may be distributed among a dozen or more elements (*44*).

Identification of the various individual nuclear reactions involved requires that each product isotope be identified both as to mass number and atomic number. Chemical separations effect a preliminary sorting of the isotopes according to the atomic number. Mass numbers may then be determined by other means, particularly by an investigation of radiation characteristics or by establishing genetic relationships to known isotopes.

If the preliminary sorting is slow, many short-lived isotopes will have decayed before this step is completed, and knowledge of the reactions involved will be incomplete. Speed in the chemical separations is therefore of great importance. Fig. 22 illustrates the principal pieces of apparatus used for rapid semimicro separation methods.

Since the apparatus employed is small and the quantities of reagents used are correspondingly small, all of the materials necessary for a number of chemical separations may be enclosed in a box roughly similar to that shown in Fig. 21. The development of boxes of this kind for handling radioactive materials is due largely to Nelson Garden and the Health Chemistry Group of the Radiation Laboratory at the University of California. If high levels of β and γ activity are to be handled, additional shielding of lead may be placed around the box. The use of lead glass windows and the substitution of mechanical manipulators for the gloves shown provide adequate protection against all but exceptionally high levels (> 0.5 curies) of β and γ activities.

Similar boxes in which a series of elaborate chemical operations may be carried out on the curie level have also

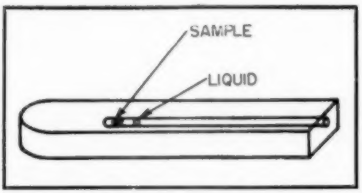


FIG. 20. Capillary cell for measuring density of very small solid samples. Top and bottom faces of cell are ground and polished to give optically flat surfaces

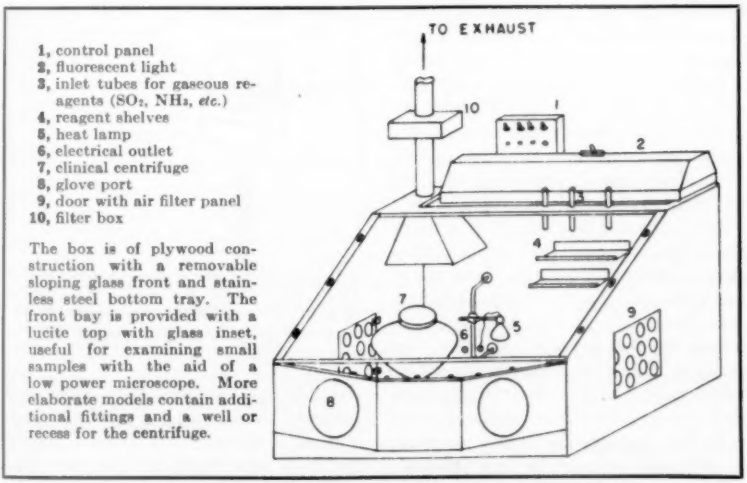
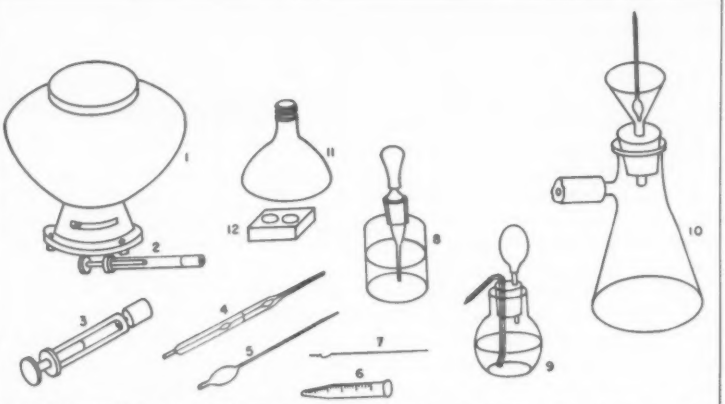

TO EXHAUST
1, control panel
2, fluorescent light
3, inlet tubes for gaseous reagents (SO_2, NH_3, *etc.*)
4, reagent shelves
5, heat lamp
6, electrical outlet
7, clinical centrifuge
8, glove port
9, door with air filter panel
10, filter box
The box is of plywood construction with a removable sloping glass front and stainless steel bottom tray. The front bay is provided with a lucite top with glass inset, useful for examining small samples with the aid of a low power microscope. More elaborate models contain additional fittings and a well or recess for the centrifuge.

FIG. 21. Box for working with radioactive materials



1, clinical centrifuge; **2,** 1-ml syringe control; **3,** 10-ml syringe fitted with rubber stopper adapter; **4,** calibrated pipet; **5,** transfer pipet; **6,** 15-ml graduated cone; **7,** platinum stirring rod; **8,** dropping bottle; **9,** 50-ml wash bottle; **10,** pipet cleaner; **11,** heat lamp; **12,** block supporting sample plates

FIG. 22. Semimicro apparatus

been designed by Mr. Garden and his group.

Microtarget for Cyclotron Bombardments

In the case of the cyclotron, and other particle-accelerating devices, the beam in the target area is considerably more concentrated at the center than at the edges. If a limited amount of material is available for bombardment, a higher specific activity may be obtained per unit of bombardment time by confining the target material to the more intense area of the beam, providing that the material so disposed still behaves as a "thin" target. The design of a micro interceptor target, based on this principle, is shown in Fig. 23. The target has been used by Thompson, Ghiorso, and Cunningham (*45*) for bombardments of the actinide elements. In practice, elaborate precautions must be taken to protect the research personnel and the cyclotron from contamination with the alpha-active target material. A successful solution to this problem, without which the bombardments referred to could not have been made, was achieved by the use of a very ingenious apparatus designed by Webb (*46*) of Dr. J. G. Hamilton's group at the Radiation Laboratory.

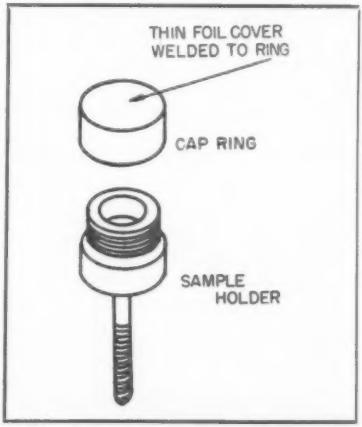


FIG. 23. Platinum micro interceptor target. During bombardment the shank of the target is screwed into a water-cooled block (not shown)

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SCIENTISTS ARE LARGELY TO BLAME for the fact that the intangible rewards of scientific investigation are not generally understood. One never hears musicians or sculptors or poets justify their role in society with the claim that they increase the physical well-being of their fellow men. Society values them, insofar as it values them at all, for the pleasure they give to life. Scientists, however, emphasize the material benefits of science because these are readily comprehended and accepted. In doing so, they misrepresent and belittle some of their major contributions to human welfare. Scientific research is one of the great adventures of the human mind. When the spirit of that adventure is generally understood, it will quicken the life and raise the hopes of people everywhere.

—*Detlev W. Bronk*, Science **109,** 478 (1949)

NUCLEONIC EVENTS

U. S., U. K., CANADA HOLD TOLERANCES CONFERENCE

A three-nation conference on radiation tolerances was held September 29–30 at Chalk River, Ontario. The conference was held under the Technical Cooperation Program of the United States, the United Kingdom, and Canada, which was established early in 1948 and does not include weapon information.

One of the primary purposes of the conference was to discuss the establishment of uniform radiation tolerance standards between the atomic energy projects of the three nations.

The eleven delegates who attended from the United States were: John Z. Bowers, Deputy Director, Div. of Biology and Medicine, AEC, Washington, D. C.; Austin M. Brues, Director, Biology Div., Argonne Natl. Lab., Chicago, Ill.; G. Failla, Prof. of Radiology, Columbia Univ. Med. School, New York; J. G. Hamilton, Director, Crocker Lab., Univ. of Calif., Berkeley, Calif.; Louis Hempelmann, Special Asst. to Director, Div. of Biology and Medicine, AEC, Washington, D. C.; Wright Langham, Los Alamos Scientific Lab., Los Alamos, N. Mex.; Karl Z. Morgan, Director, Health Physics Dept., Oak Ridge Natl. Lab., Oak Ridge, Tenn.; Herbert M. Parker, Manager, Health Instruments Div., Hanford Plutonium Works, Hanford, Wash.; Lauriston S. Taylor, Consultant to the Biophysics Branch, AEC, Washington, D. C.

Prior to the conference, three of the British delegates visited several AEC installations to discuss various health and safety matters. They were: Joseph S. Mitchell, Cambridge Univ.; Gerard J. Neary, Medical Research Council, Biological Unit, Harwell, England; and Arthur C. Chamberlain, Atomic Energy Research Establishment, Harwell.

AFRICAN AIRWAYS FLY RADIOISOTOPES IN WING-TIPS

A technique has been developed for transporting radioisotopes cheaply in the wing-tips of Skymasters used by South African Airways between the Union and the United Kingdom. The method eliminates the heavy lead casings that were previously necessary to protect crews and freight against radiation.

The isotopes are enclosed in small metallic containers fitted with a ring into which a metal rod is hooked for loading into the wing-tips.

Radiocobalt was placed in a wing-tip container of a Skymaster for 24 hours, and 20 X-ray plates were distributed in the pilot's cabin, passengers' cabin, and freight bags. Exhaustive tests showed that gamma rays from the cobalt did not affect crews, passengers, or photographic plates carried as air freight.

AEC APPROVES 18 PROPOSALS FOR LIFE SCIENCES RESEARCH

Negotiations leading to the award of contracts for 18 research proposals in the field of biology and medicine are underway, the Atomic Energy Commission recently announced.

The award of contracts to the 18 institutions involved brings to a total of 150 the number of AEC-supported research projects being carried on in medicine, biology, cancer studies and biophysics at universities, hospitals

and research centers. Approximately $5,000,000 has been earmarked by the AEC for support of such research in non-government agencies during fiscal year 1950.

The institutions which submitted the newly-approved proposals are listed below. The AEC negotiating authority is noted in each case in italics.

Medicine

Massachusetts General Hospital (Drs. William M. Sweet and Bertram Selverstone): For use of phosphorus-32 in precise localization of brain tumors. *New York Operations Office.*

University of Oregon Medical School (Dr. Edward E. West): For study of labeled acetic acid and ethanol in relation to fat metabolism. *Hanford Operations Office.*

University of Tennessee (Dr. E. F. Williams): For research in pathology, physiology and chemistry. *Oak Ridge Operations Office.*

Presbyterian Hospital of the City of Chicago (Dr. R. Gordon Gould): For study of the mechanism of CO_2 fixation. *Chicago Operations Office.*

Peter Bent Brigham Hospital (Dr. Francis D. Moore): For study of intracellular changes in trauma depletion and repair; and biochemical studies in the human being with the aid of isotopes. *New York Operations Office.*

University of Denver (Dr. Frank M. D'Amour): For study of physiologic and pathologic effects of radioactive cobalt. *Santa Fe Operations Office.*

Harvard University (Dr. Thomas H. Ham and Dr. William B. Castle): For study of destruction of red blood cells. *New York Operations Office.*

Harvard University (Dr. S. P. Hicks): For study of effects of radiation upon development of rat embryos. *New York Operations Office.*

Syracuse University (Dr. E. L. Lozner): For study of defenses against hemorrhages. *New York Operations Office.*

Washington University, St. Louis (Dr. Frank Dixon): For investigation of effects of agents used in treatment of cancer; and study of X-ray and nitrogen mustards on immunologic response of experimental animals. *Chicago Operations Office.*

University of Illinois (Dr. A. C. Ivy): For irradiation of gastric mucosa by intragastric instillation of radioactive isotopes. *Chicago Operations Office.*

Biology

Iowa State College (Dr. S. Aranoff): For study of metabolism and physiology of roots, and (Dr. R. E. Sealock): For study of combined biochemical and physiological action of throsine and vitamin B_{12}. *Chicago Operations Office.*

Agricultural and Mechanical College of Texas (Dr. Raymond Reiser and Dr. Kenneth Kuiken): For study of metabolism of glycerines. *Oak Ridge Operations Office.*

University of Pennsylvania (Dr. D. Wright Wilson): For study of synthesis of isotopic carbon compounds used in biochemistry. *New York Operations Office.*

University of Chicago (Dr. E. M. K. Geiling): For study of biosynthesis of radioactive drug compounds. *Chicago Operations Office.*

University of Wisconsin Agriculture Experiment Station (Drs. B. W. Burris and P. W. Wilson): For studies of biological nitrogen fixation with isotopic tracers; and study of metabolism of organic acids in higher plants and microorganisms. *Chicago Operations Office.*

Applied Biophysics

University of Washington (Dr. F. W. Church and Dr. Raymond Allen): For meteorological studies. *Hanford Operations Office.*

University of Washington (Dr. Robert G. Fleagle): For meteorological studies. *Hanford Operations Office.*

NEW BRANCH CHIEFS NAMED BY AEC

The Atomic Energy Commission has announced the appointment of two new branch chiefs to its Washington headquarters staff. Dr. Walter D. Claus, formerly head of the physical-chemical division of the Pabst Research Laboratories, Milwaukee, Wisc., has been named as chief of the Biophysics Branch, Division of Biology and Medicine.

Dr. Joseph B. Platt has been named chief of the Physics Branch, Division of Research, a new office established to administer the AEC research program in the field of physics. Dr. Platt formerly was an associate professor at the University of Rochester.

Dr. Claus replaces Dr. Lauriston S. Taylor, who has returned to the position of chief of the X-ray Section at the National Bureau of Standards, Washington, D. C. Dr. Taylor had been on loan from the Bureau to the Commission for the past year. He has been retained by the AEC as a consultant on radiation matters.

ELEMENTARY PARTICLES DISCUSSED AT U. OF EDINBURGH

On November 14–16, the University of Edinburgh was host to scientists attending a Conference on Elementary Particles. Among the papers presented were:

"Dies Erzeugung von Mesonen in Vielfachprozessen," W. Heisenberg; "On the production of mesons by nucleons," L. Janossy; "Quantum electrodynamics and correspondence-principle," H. A. Kramers; "Some new measurements on the nature of the vertical cosmic ray

beam at sea level," J. G. Wilson; "General theory of elementary particles," M. Born; "Experimental evidence concerning the possible existence of the negative proton and the di-neutron," N. Feather; "Reciprocity theory of electrodynamics," K. C. Cheng.

Other participants included F. Bopp, M. Fierz, L. Leprince-Ringuet, C. Moller, C. F. Powell, A. Proca, and H. O. W. Richardson.

SANDIA CORP. CREATED TO OPERATE LAB

Western Electric Co., which has signed a contract with the AEC for the operation of Sandia Laboratory, Albuquerque, N. M., will run the laboratory through a new, wholly-owned subsidiary named the Sandia Corporation. The new organization will call upon Western Electric Co. and the Bell Telephone Laboratories, both subsidiaries of the American Telephone and Telegraph Co., for scientific, technical, and managerial services and assistance.

The laboratory was transferred from the present operator, the University of California, to the Western Electric Co. on November 1, 1949. The new contract is effective through December 31, 1953.

AEC, NOTRE DAME SPONSOR NEW ATOM SMASHER

A 2,000,000-volt electrostatic generator which will produce high-velocity electrons and X-rays will be constructed on the South Bend, Ind., campus of the University of Notre Dame, Rev. John J. Cavanaugh, president of the university, has announced. Under the joint sponsorship of the Atomic Energy Commission and the university, the generator will be housed in the $1,400,000 Science Building scheduled for construction next spring. The new machine, second to be built at Notre Dame, will be used in experiments on the effects of radiation.

NEW ELEMENT NAMES OFFICIALLY ADOPTED

At a meeting of the American Chemical Society held in Atlantic City, N. J., September 23, the official nomenclature for eight new chemical elements adopted at a recent meeting of the International Union of Chemistry was announced.

The four transuranium elements received the names proposed by their discoverers, as follows:

Element	*Officially named*
93	Neptunium
94	Plutonium
95	Americium
96	Curium

Four other elements, the nomenclature for which has caused considerable confusion in the past, have been named as follows:

Element	*Officially named*	*Formerly known as*
43	Technetium	Masurium
61	Promethium	Illinium
85	Astatine	Virginium
87	Francium	Alabamine

Other names officially adopted were:

4	Beryllium	Glucinium
41	Niobium	Columbium
71	Lutetium	Lutecium
72	Hafnium	
74	Wolfram	Tungsten
91	Protactinium	Protoactinium

HEALTH SERVICE PUBLISHES DONORA DISASTER REPORT

The Public Health Service recently released its 173-page report summarizing six months of on-the-spot study by 35 medical, chemical, engineering, and meteorological experts of the week-long smog in October, 1948, at Donora, Pa., which was accompanied by the death of some 20 persons.

No one chemical compound found in the air, nor no combination of chemical pollutants and particles of solid matter

was found to be the cause of the deaths. Instead, Public Health Service could only report, "It seems reasonable to state . . . that (the disaster) could have been produced by a combination, or summation of the action of two or more of the contaminants."

20 PAPERS PRESENTED AT IRE-AIEE CONFERENCE

Twenty papers were presented at the Second Annual Joint IRE-AIEE Conference on Electronic Instrumentation in Nucleonics and Medicine held October 31–November 2 in New York. In addition, a special session featured a round-table discussion of radiation hazards with J. R. Dunning of Columbia University as moderator for the four-man panel of participants which included A. L. Baker, Kellex Corp.; Ralph E. Lapp, author of "Must We Hide"; Adm. W. S. Parsons, Dept. of Defense; and Shields Warren, director of the AEC's Division of Biology and Medicine. An address, following the round-table discussion, was delivered by Lewis L. Strauss, member of the AEC.

OAK RIDGE OFFERS ADDITIONAL RADIOTRACER COURSES

As a continuation of a series of courses offered during the past year and a half, three additional basic courses in the techniques of using radioisotopes as tracers are being offered this winter by the Special Training Division of the Oak Ridge Institute of Nuclear Studies.

The dates the courses will be held are January 2–27, January 30–February 25, and March 6–31, 1950.

Each session will be divided into laboratory work, lectures on laboratory experiments, general background lectures, and special-topic seminars. Experiments will be conducted covering the use and calibration of instruments and the purification and separation of radioactive materials from inert materials and from other radioactive materials. Other laboratory work will be devoted to the application of various radioisotope techniques.

Thirty-two participants will be accepted for each course. Application for the first course should be mailed prior to December 1. A registration fee of $25 is charged for each participant. Application forms and additional information may be obtained from R. T. Overman, Special Training Division, Oak Ridge Institute of Nuclear Studies, P. O. Box 117, Oak Ridge, Tenn.

NUCLEAR NEWSMAKERS

William Mason has been appointed as head of the Bacteriology and Virology Division at Brookhaven National Laboratory. **Sidney C. Madden** has been appointed to head the Pathology Division of Brookhaven's Department of Medicine.

Jesse C. Johnson has been appointed to the newly established position of Deputy Manager, AEC Raw Materials Operations Office, Washington, D. C. He had formerly served as assistant manager of the same office, in charge of domestic production.

Michael Amrine, formerly head of Public Education at Brookhaven National Laboratory, has been signed by Bell Syndicate, Inc., to write "You and the Atom," a newspaper column dedicated entirely to atomic energy and its impact on national and foreign affairs.

T. H. Johnson, the chairman of the Physics Department, Brookhaven National Laboratory, last month visited the Canadian Atomic Energy Project at Chalk River to discuss the use of nuclear reactors for physical research.

David E. Lilienthal, AEC'S chairman, and Gen. Lucius E. Clay, received the 1949 Freedom Awards from Freedom

House at a dinner in New York on October 13. Lilienthal was presented with a plaque inscribed "Pathfinder of a new age harnessing nature and science in the service of democracy."

Thomas B. Lanahan, III, has joined S. Blickman, Inc., as specialist on equipment for handling radioactive materials.

Carl F. von Weizsäcker, German scientist who was the first to propose a detailed system of nuclear reactions to account for the source of energy in the sun and stars, is presenting a series of public lectures on "The Modern Concept of Nature" at the University of Chicago and will teach there as Alexander White visiting professor this term.

HYMAN H. GOLDSMITH (1907–1949)

Hyman H. Goldsmith served as a valued adviser to the editor of NUCLEONICS during the first few and critical months of its existence. Both his early assistance and the help he gave in the later months of his life were important and timely for the success of this international journal.

—Norman R. Beers

In the untimely death of Hyman H. Goldsmith, American physics has suffered a serious loss. Teacher, research worker, encyclopedist, editor, student of the social implications of science—Goldsmith's career closely paralleled the growth of nuclear physics in this country, and a rare vision enabled him to comprehend the important role that physics and physicists were to play in the post-war world.

He started teaching physics at C.C.N.Y. in his senior year, and continued until World War II. His classes were seldom conventional and never dull, for Goldsmith had no taste for drudgery, and that which could easily be learned from the textbook was of little interest to him. To the good student his courses were a joy, and in them many a young physicist was introduced to art, classical and modern music, philosophy, and nuclear physics.

One of Goldsmith's lectures stands out clearly in my mind. It was in 1936 in my freshman "Science Survey" course. The neutron had only recently been discovered; its properties were just then being studied. At Columbia, Goldsmith was investigating the interactions of slow neutrons with nuclei, and this day he brought to class a radon-beryllium neutron source and a gallon jug of water. In the course of the lecture, he activated a silver half-dollar with slow neutrons and demonstrated its radioactive properties. To me, as a beginning student of physics, this demonstration made the subject come alive.

Already, in this (prewar) period, Goldsmith's talent as an encyclopedist of nuclear physics was well known. He could deliver, and was frequently called upon to deliver, a colloquium on any one of the rapidly developing aspects of nuclear physics or cosmic ray research. He was a "walking handbook" of information on nuclear physics. But his unique quality as a source of information lay in his critical judgment, in his ability to discriminate between good and poor experiments, and between significant and trivial results.

When the war came, Goldsmith joined the Manhattan Project, as head of the Physics Information branch of the Metallurgical Laboratory, at the

University of Chicago. He helped to organize and develop the "Chicago Handbook" of nuclear data, which was so useful throughout the Manhattan Project. His famous collection of cross section curves—known within the "project" as the "wallpaper book" (later published, in a declassified version, in the *Reviews of Modern Physics* as "Neutron Cross Sections of the Elements")—set a high standard of intelligent and useful data compilation.

To most of the men who had worked on the bomb project, the end of the war signaled the beginning of a new and more vital task: the achievement of a system of effective world control of atomic energy and other potential weapons of mass destruction. Goldsmith threw himself—with characteristic energy and devotion—into the organization of the Federation of Atomic Scientists and in the political fight for the principle of civilian control of atomic energy.

But Goldsmith's major contribution to the Atomic Scientist movement was in the founding and co-editorship of the *Bulletin of the Atomic Scientists* (of which Eugene Rabinowitch has written on the next page). He had high professional standards, and amateurism in social and political science was as abhorrent to him as was a slipshod experiment in nuclear physics. He inspired and obtained articles by the best minds in many fields. He loved a good argument, and was scrupulously fair about publishing all sides, so that many a lively controversy was carried on in the *Bulletin's* pages. His use of the *Bulletin*, for the early publication of the titles of "declassified" documents, helped a great deal towards clarifying and liberalizing the Manhattan District declassification procedures.

In 1947 he became head of the Information and Publication Division of the newly organized Brookhaven National Laboratory. In this capacity, and also as Chairman of the Subcommittee on Publications of the Committee on Nuclear Science of the National Research Council, he strove to establish and maintain a high professional standard of data compilation and publication. The compilations of nuclear moments and nuclear energy levels, assembled and published at Brookhaven, were the first fruits of a project designed to cover all of nuclear physics, and subsequently, other branches of nuclear science. His weekly summary of the current literature and his guide to Russian Scientific Literature were widely disseminated and extensively used.

Goldsmith's interests were so diverse, and his energy so prodigious, that such a summary can only include a fraction of his activities. At the time of his death, he was engaged in the early stages of preparation of a complete "Handbook of Nuclear Physics." Just prior to his death he spent a few weeks in Paris, where he participated as a representative of U.N.E.S.C.O., in an international conference on science abstracting.

These paragraphs would be incomplete without a few comments concerning the qualities of Hy as a person. I had the good fortune and the pleasure of knowing him well, in work and in play, over a period of almost fifteen years. He was a devoted friend, and while he often assumed the role of severe (albeit scrupulously fair) critic, he always understood and usually excused the particular weakness which called forth the criticism. The days we spent together in Paris, just before his return to the United States and his unfortunate accident, were full of delightful experiences and insights. His children found him a delightful companion, for his reactions had a rare freshness and spontaneity, and he was, to them, a friend and playfellow, as well as a father. Those of us who had the good fortune to know him as a friend will not soon forget him, or cease to mourn over his untimely departure.

—Bernard T. Feld, Massachusetts Institute of Technology

Hy Goldsmith had a very wide range of knowledge and extensive interests—physics, politics, arts, and people. This was perhaps why he was never able to

settle down to belaboring one small corner in the wide field of science, as is the fate of most scientists, except some very great or very fortunate ones. This wide interest finally determined his choice of profession as head of Brookhaven's Information Division.

I worked with Hy Goldsmith for five years, one year in the Information Division of the Metallurgical Laboratory, and four years as co-editor of the *Bulletin of the Atomic Scientists*.

Although Brookhaven took most of his time and was the source of his living in the last three years of his life, I think it is fair to say that the activity nearest his heart during this time was the *Bulletin*. In 1945 when the "Atomic Scientists" first burst into the limelight, and became familiar figures in public meetings, and dining rooms of Washington hostesses, Goldsmith conceived the idea of a magazine that would assist the scientists in their political fight for domestic civilian rule, and for international control of atomic energy. After a few discussions of this project in our favorite corner drug store, Hy Goldsmith went ahead. In his characteristic impatient and optimistic way, he secured, from an obscure printer in West Chicago, enough paper for 500 copies, and before anyone had time to think twice whether the venture was justified, the first issue of the *Bulletin* was off the press (on December 15, 1945). It was planned to be four pages long; but Hy cheerfully exceeded the appropriation allotted by the Atomic Scientists of Chicago, and made it six. This established a pattern for the *Bulletin's* growth.

For Hy Goldsmith, the *Bulletin* was the most important thing on earth. In Editorial Board meetings, he tolerated no pessimism, defeatism, or pusillanimity. His optimism has carried the *Bulletin* through the many financial and psychological crises of the past four years.

He did not himself write for the *Bulletin*. He was too much a man of action to sit down at the desk long enough to produce an article. Moreover, he was unenthusiastic about accepting articles only because their authors were personally or professionally closely associated with the "atomic scientists." In his mind, the most famous writers were just good enough and should be proud of having their articles published in the *Bulletin*. Despite his irregular participation in the *Bulletin* affairs (he was always rushing across the country, and it was often difficult for the office staff to locate him), he jealously guarded his right to read, criticize, and/or reject every article or short note submitted. In his criticism of vagueness, triteness, smugness, or sentimentalism, he had no consideration for the author's importance or personal associations. Here, as in all his dealings, he enjoyed rude frankness. No colleague, however famous or superior in rank, was secure from his verbal lashings. Some kept a grudge against him for a long time, but the majority remained his friends. Despite his bitter—and occasionally unjust—attacks on those who did not live up to his expectations, there was something eminently likable about Hy Goldsmith.

His untimely death was characteristic of him—his fatal fall, too, was the result of acting on an impulse, walking upright over a dangerous slippery place across a waterfall with a cramp in his leg, instead of resting and then carefully negotiating the dangerous passage. He leaves a sizable contribution to American scientific documentation in the form of the Brookhaven library and associated abstracting and translating services, and in the collection of nuclear data prepared by him; but I am certain that he will be remembered mainly by his burning interest in bringing together physical and social science, in kindling the feeling of political responsibility in natural scientists and making non-scientists aware of the implications of science in world affairs.

—Eugene Rabinowitch, Editor
Bulletin of the Atomic Scientists

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REFLECTIONS

This space will be used to publish, with the permission of the author, communications received in correspondence when the content is technical and believed to be of interest to our readers.

DEAR SIR:

Dr. K. J. McCallum, in his excellent article in the July NUCLEONICS (p. 11), described the technique of preparing radium-beryllium neutron sources in considerable detail, and may have thereby encouraged chemists and physicists to undertake the preparation of their own sources. While he did put in a word of warning, it seemed to me that the radiation and contamination hazards involved in the manipulation of radium were not sufficiently emphasized.

From personal experience in this field I would advise strongly that such work should not be undertaken in any laboratory that is not fully equipped and experienced in the handling of radioactive materials of long life and high specific activity. Full consideration should be given to the gamma radiation intensities involved, to the consequences of a "spill," to the possibility of contamination of the personnel and the laboratory, and to the publicized toxicity of beryllium, before the preparation of such sources is undertaken.

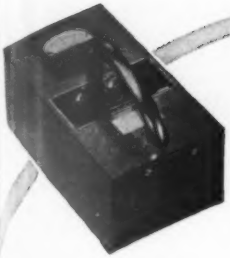
ADAIR MORRISON

Physics Division
National Research Council of Canada
Ottawa, Ontario

DEAR SIR:

The editorial you composed (NUCLEONICS, July, p. 2) includes some provocative points. I was particularly stimulated by the thoughts expressed in the

(Continued on page 96)



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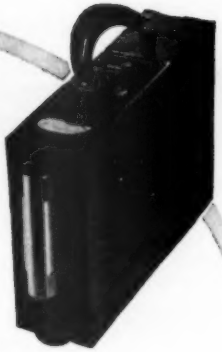

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"Reflections" (Continued from page 94)

last paragraph which suggest that unless extensive measurements of human exposure to radiation throughout the country are started soon, we shall be in the same pickle thirty or forty years from now with respect to evaluating tolerances as we find ourselves in the problem of silicosis and other long-developing occupational diseases. Lack of information on exposures of years ago have greatly impeded the development of exposure standards today.

W. C. L. HEMEON

Director
Industrial Hygiene Foundation of America
Pittsburgh, Pennsylvania